

09/787231

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Docket No. E-6893  
 Date March 16, 2001

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THE ASSISTANT COMMISSIONER FOR PATENTS  
 Washington, D. C. 20231

[ ] ATTN: BOX PATENT APPLICATION  
 [X] ATTN: BOX PCT

**[X] THIS IS THE NATIONAL STAGE OF PCT/JP00/05812 FILED August 28, 2000**

Sir:

Transmitted herewith for filing is the [X] Utility [ ] Design patent application of:

Inventor/Application Identifier: **Masahito NISHIURA et al.**

For: **POLYMERIC ELECTROLYTE, NONAQUEOUS ELECTROLYTE AND ELECTRIC DEVICE USING THE SAME**

Enclosed are:

[ ] \_\_\_\_\_ sheets of drawings ([ ] formal [ ] informal size A4).

[X] 45 pages of specification, including claims and abstract.

[X] 45 total pages

[ ] Combined Declaration/Power of Attorney

[ ] Newly executed

[ ] Copy from prior application

[ ] Inventors deleted; see attached statement

[X] Inventor Information Sheet

[ ] Incorporation By Reference. The entire disclosure of the prior application, from which a copy of the oath or declaration is supplied, is considered as being part of the disclosure of the accompanying application and is hereby incorporated by reference therein

[ ] Sequence Listing

[ ] Computer Readable Copy

[ ] Paper copy

[ ] The undersigned hereby affirms that the content of the paper and computer readable copies of the Sequence Listing are the same.

[ ] Cancel in this application original claims \_\_\_\_\_ of the prior application before calculating the filing fee.

#### CLAIMS FILED

For	Number Filed	Number Extra	Rate	Basic Fee
Total Claims	65	45 (over 20)	x \$18.00	\$810.00
Independent Claims	2	0 (over 3)	x \$80.00	
[X] Multiple Dependent Claim			\$270.00	270.00
[ ] Reduce by 50% for Small Entity				
[ ] Foreign Language Filing Fee			\$130.00	
<b>TOTAL FILING FEE</b>				<b>\$1,940.00</b>

[X] Please charge Deposit Account No. 10-1250 in the amount of  
 A duplicate copy of this sheet is attached.

**\$1,940.00**

[X] Please charge to Deposit Account No. 10-1250 any further fees under  
 37 CFR 1.16; 37 CFR 1.17; 37 CFR 1.492.

[ X ] Return Receipt Postcard

[ X ] Preliminary Amendment

[ ] Assignment to \_\_\_\_\_

[ ] Assignment is of record in prior application Serial No. \_\_\_\_\_.

[ ] Assignment Recordation Form Cover Sheet.

[ ] Charge \$40.00 to Deposit Account No. 10-1250 for recording Assignment.

[ ] Information Disclosure Statement and/or Information Disclosure Citation

[ ] English translation

[ ] Small Entity Status is asserted

[ ] Applicant hereby claims the benefit of the filing date of the following provisional application(s) under the provisions of 35 USC 119.

[ X ] Applicant hereby claims the benefit of the filing date of the following applications under the provisions of 35 USC 119 of which certified copies [ ] will follow [ ] are enclosed [ X ] have been filed in the International Bureau [ ] were filed in prior application No. \_\_\_\_\_.

(1) Japan Appln. No. 11-248890, filed September 2, 1999

(2) Japan Appln. No. 11-248891, filed September 2, 1999

[ ] This is a [ ] Continuation [ ] Divisional [ ] Continuation-in-Part of prior application Serial No. \_\_\_\_\_.

[ ] Amend the specification by inserting before the first line the sentence:

--This is a [ ] continuation, [ ] division, [ ] continuation-in-part, of application Serial No. , filed . --

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F-6893

Inventor Information Sheet

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Nationality : Japan

F-6893

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant : Masahito NISHIURA et al.  
Serial No. : Not yet known (U.S. National Stage of  
PCT/JP00/05812 filed August 28, 2000)  
For : POLYMERIC ELECTROLYTE, NONAQUEOUS  
ELECTROLYTE AND ELECTRIC DEVICE  
USING THE SAME

Assistant Commissioner for Patents  
Washington, D.C. 20231

**PRELIMINARY AMENDMENT**

Sir:

Preliminary to examination, please amend this application as follows:

**IN THE CLAIMS**

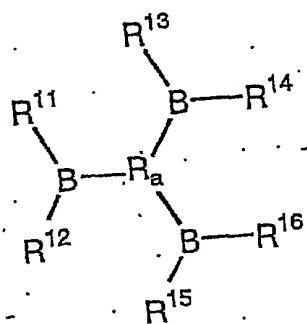
Amend claims 1-27 as follows:

1. (Amended) A polymeric electrolyte comprising an electrolytic salt, a polymeric compound forming a complex with the electrolytic salt, and a compound having boron atoms.
  
2. (Amended) The polymeric electrolyte according to claim 1, wherein the compound having boron atoms is one or more selected from the group

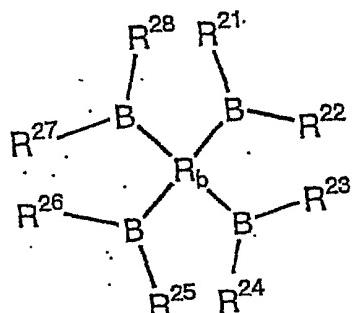
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consisting of compounds represented by the following general formulas (1) to (4)

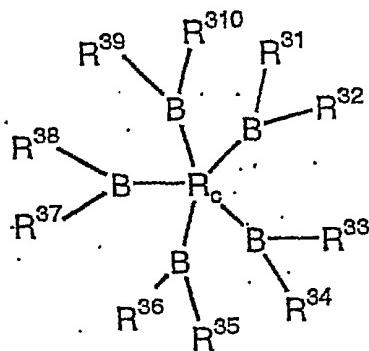
general formula (1)



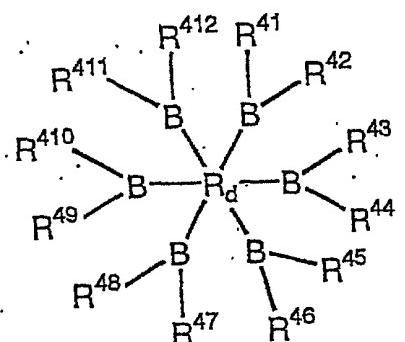
general formula (2)



general formula (3)



general formula (4)



wherein

$R^{11}, R^{12}, R^{13}, R^{14}, R^{15}$  and  $R^{16}$  in formula (1),  $R^{21}, R^{22}, R^{23}, R^{24}, R^{25}, R^{26}, R^{27}$  and  $R^{28}$  in formula (2),  $R^{31}, R^{32}, R^{33}, R^{34}, R^{35}, R^{36}, R^{37}, R^{38}, R^{39}$  and  $R^{310}$  in formula (3), and  $R^{41}, R^{42}, R^{43}, R^{44}, R^{45}, R^{46}, R^{47}, R^{48}, R^{49}, R^{410}, R^{411}$  and  $R^{412}$  in formula (4), which may be the same or different, each represent a hydrogen atom, a halogen atom or a monovalent group, or are bound to each other to form a ring,

$R_a$  in formula (1) represents a group having a site capable of being bound to at least 3 boron atoms which are the same or different,  $R_b$  in formula (2) represents a group having a site capable of being bound to at least 4 boron atoms which are the same or different,  $R_c$  in formula (3) represents a group having a site capable of being bound to at least 5 boron atoms which are the same or different, and  $R_d$  in formula (4) represents a group having a site capable of being bound to at least 6 boron atoms which are the same or different.

3. (Amended) The polymeric electrolyte according to claim 2, wherein  $R^{11}, R^{12}, R^{13}, R^{14}, R^{15}, R^{16}, R^{21}, R^{22}, R^{23}, R^{24}, R^{25}, R^{26}, R^{27}, R^{28}, R^{31}, R^{32}, R^{33}, R^{34}, R^{35}, R^{36}, R^{37}, R^{38}, R^{39}, R^{310}, R^{41}, R^{42}, R^{43}, R^{44}, R^{45}, R^{46}, R^{47}, R^{48}, R^{49}, R^{410}, R^{411}$  and  $R^{412}$  are one or more of an alkyl group, an aryl group and flourine- substituted derivatives thereof.

4. (Amended) The polymeric electrolyte according to any one of claims 1 to 3, wherein the polymeric compound is one or more selected from the

group consisting of a polyalkylene, a polyether, a polyester, a polyamine, a polyimide, a polyurethane, a polysulfide, a polyphosphazene, a polysiloxane, derivatives thereof, copolymers thereof and crosslinked products thereof.

5. (Amended) The polymeric electrolyte according to any one of claims 1 to 3, wherein the polymeric compound is one or more selected from the group consisting of a polyalkylene oxide, polyvinylidene fluoride, polyhexafluoropropylene, polyacrylonitrile, polymethyl methacrylate, derivatives thereof, copolymers thereof and crosslinked products thereof.

6. (Amended) The polymeric electrolyte according to any one of claims 1 to 3, wherein the electrolytic salt is a metallic salt.

7. (Amended) The polymeric electrolyte according to claim 6, wherein the metallic salt is a lithium salt.

8. (Amended) The polymeric electrolyte according to claim 7, wherein the lithium salt is one or more selected from the group consisting of LiBF<sub>4</sub>, LiPF<sub>6</sub>, LiClO<sub>4</sub>, LiAsF<sub>6</sub>, LiCF<sub>3</sub>SO<sub>3</sub>, LiN(CF<sub>3</sub>SO<sub>2</sub>)<sub>2</sub>, LiN(C<sub>2</sub>F<sub>5</sub>SO<sub>2</sub>)<sub>2</sub>, LiC(CF<sub>3</sub>SO<sub>2</sub>)<sub>3</sub>, LiCl, LiF, LiBr, LiI, derivatives thereof and like.

9. (Amended) The polymeric electrolyte according to any one of claims 1 to 3, further comprising a nonaqueous solvent.

10. (Amended) The polymeric electrolyte according to claim 9, wherein the nonaqueous solvent is an aprotic solvent.

11. (Amended) The polymeric electrolyte according to claim 10, wherein the aprotic solvent is one or more selected from the group consisting of carbonates, lactones, ethers, sulfolanes and dioxolanes.

12. (Amended) The polymeric electrolyte according to any one of claims 1 to 3, wherein the molar ratio of the compound having boron atoms to the electrolytic salt is 0.1:100 to 300:100.

13. (Amended) An electric device comprising the polymeric electrolyte according to any one of claims 1 to 3.

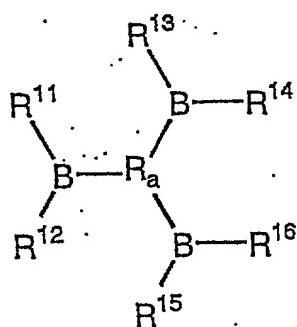
14. (Amended) A cell comprising a positive electrode, a negative electrode and the polymeric electrolyte according to any one of claims 1 to 3, said electrodes being linked through said electrolyte.

15. (Amended) The cell according to claim 14, wherein the positive electrode is made of a double metal oxide capable of occluding and releasing lithium ions, and the negative electrode is made of a lithium metal, a lithium alloy or a compound capable of reversibly occluding and releasing lithium ions.

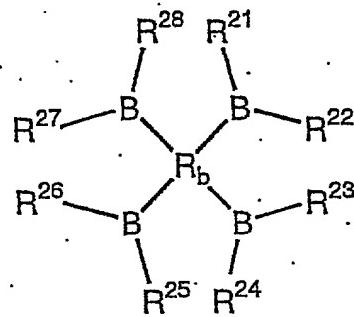
16. (Amended) A nonaqueous electrolyte comprising an electrolytic salt, a nonaqueous solvent that dissolves the electrolytic salt, and a compound having boron atoms.

17. (Amended) The nonaqueous electrolyte according to claim 16, wherein the compound having boron atoms is one or more selected from the group consisting of compounds represented by the following general formulas (1) to (4)

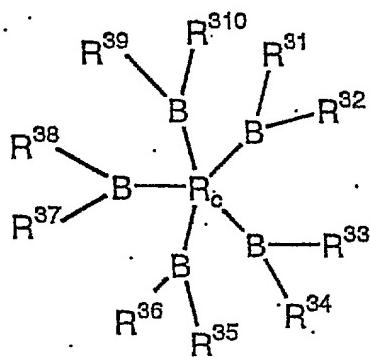
general formula (1)



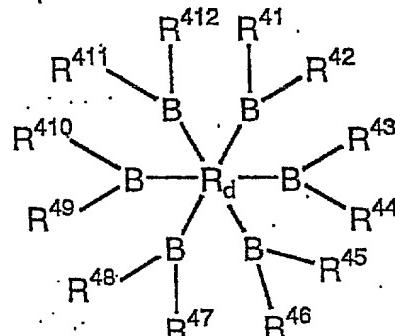
general formula (2)



general formula (3)



general formula (4)



wherein

$R^{11}, R^{12}, R^{13}, R^{14}, R^{15}$  and  $R^{16}$  in formula (1),  $R^{21}, R^{22}, R^{23}, R^{24}, R^{25}, R^{26}, R^{27}$  and  $R^{28}$  in formula (2),  $R^{31}, R^{32}, R^{33}, R^{34}, R^{35}, R^{36}, R^{37}, R^{38}, R^{39}$  and  $R^{310}$  in formula (3), and  $R^{41}, R^{42}, R^{43}, R^{44}, R^{45}, R^{46}, R^{47}, R^{48}, R^{49}, R^{410}, R^{411}$  and  $R^{412}$  in formula (4), which may be the same or different, each represent a hydrogen atom, a halogen atom or a monovalent group, or are bound to each other to form a ring,  
Ra in formula (1) represents a group having a site capable of being bound to at least 3 boron atoms which are the same or different, Rb in formula (2) represents a group having a site capable of being bound to at least 4 boron atoms which are the same or different, Rc in formula (3) represents a group having a site capable of being bound to at least 5 boron atoms which are the same or different, and Rd in formula (4) represents a group having a site capable of being bound to at least 6 boron atoms which are the same or different.

18. (Amended) The nonaqueous electrolyte according to claim 17, wherein  $R^{11}, R^{12}, R^{13}, R^{14}, R^{15}, R^{16}, R^{21}, R^{22}, R^{23}, R^{24}, R^{25}, R^{26}, R^{27}, R^{28}, R^{31}, R^{32}, R^{33}, R^{34}, R^{35}, R^{36}, R^{37}, R^{38}, R^{39}, R^{310}, R^{41}, R^{42}, R^{43}, R^{44}, R^{45}, R^{46}, R^{47}, R^{48}, R^{49}, R^{410}, R^{411}$  and  $R^{412}$  are one or more of an alkyl group, an aryl group and fluorine-substituted derivatives thereof.

19. (Amended) The nonaqueous electrolyte according to any one of claims 16 to 18, wherein the electrolytic salt is a metallic salt.

20. (Amended) The nonaqueous electrolyte according to claim 19, wherein the metallic salt is a lithium salt.

21. (Amended) The nonaqueous electrolyte according to claim 20, wherein the lithium salt is one or more selected from the group consisting of LiBF<sub>4</sub>, LiPF<sub>6</sub>, LiClO<sub>4</sub>, LiAsF<sub>6</sub>, LiCF<sub>3</sub>SO<sub>3</sub>, LiN(CF<sub>3</sub>SO<sub>2</sub>)<sub>2</sub>, LiN(C<sub>2</sub>F<sub>5</sub>SO<sub>2</sub>)<sub>2</sub>, LiC(CF<sub>3</sub>SO<sub>2</sub>)<sub>3</sub>, LiCl, LiF, LiBr, LiI, derivatives thereof and the like.

22. (Amended) The nonaqueous electrolyte according to any one of claims 16 to 18, characterized in that the nonaqueous solvent is an aprotic solvent.

23. (Amended) The nonaqueous electrolyte according to claim 22, wherein the aprotic solvent is one or more selected from the group consisting of carbonates, lactones, ethers, sulfolanes and dioxolanes.

24. (Amended) The nonaqueous electrolyte according to any one of claims 16 to 18, wherein the molar ratio of the compound having the boron atoms in the structure to the electrolytic salt is 0.1:100 to 300:100.

25. (Amended) An electric device comprising the nonaqueous electrolyte according to any one of claims 16 to 18.

26. (Amended) A cell comprising a positive electrode, a negative electrode, the nonaqueous electrolyte according to any one of claims 16 to 18 and a separator, said electrodes being linked through said electrolytes and said separator.

27. (Amended) The cell according to claim 26, wherein the positive electrode is made of a double metal oxide capable of occluding and releasing lithium ions, and the negative electrode is made of a lithium metal, a lithium alloy or a compound capable of reversibly occluding and releasing lithium ions.

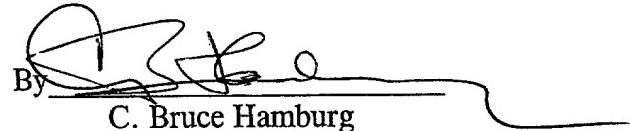
The changes are shown in the Appendix hereto.

Remarks

This places the application in better condition for examination.

Respectfully submitted,

Jordan and Hamburg LLP

  
By \_\_\_\_\_  
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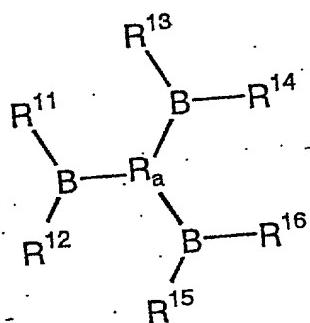
enc: Appendix

## APPENDIX

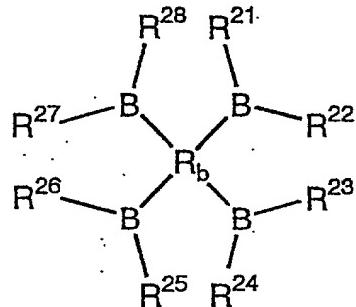
## PENDING CLAIMS WITH AMENDMENTS EFFECTED THEREIN

1. (Amended) A polymeric electrolyte [made of] comprising an electrolytic salt, [and] a polymeric compound forming a complex with the electrolytic salt, [characterized by containing] and a compound having boron atoms[ in the structure].
2. (Amended) The polymeric electrolyte according to claim 1, [characterized in that] wherein the compound having boron atoms [in the structure] is one or more selected from the group consisting of compounds represented by the following general formulas (1) to (4)[.]

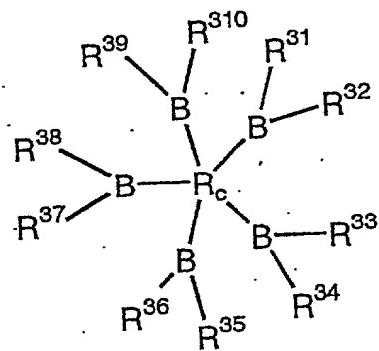
general formula (1)



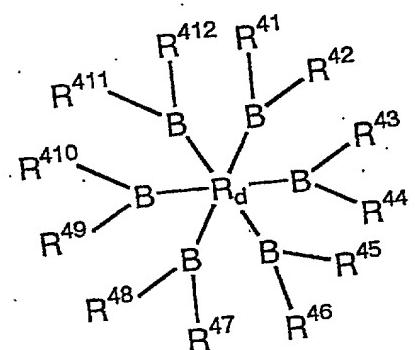
general formula (2)



general formula (3)



general formula (4)



wherein

$R^{11}, R^{12}, R^{13}, R^{14}, R^{15}$  and  $R^{16}$  in formula (1),  $R^{21}, R^{22}, R^{23}, R^{24}, R^{25}, R^{26}, R^{27}$  and  $R^{28}$  in formula (2),  $R^{31}, R^{32}, R^{33}, R^{34}, R^{35}, R^{36}, R^{37}, R^{38}, R^{39}$  and  $R^{310}$  in formula (3), and  $R^{41}, R^{42}, R^{43}, R^{44}, R^{45}, R^{46}, R^{47}, R^{48}, R^{49}, R^{410}, R^{411}$  and  $R^{412}$  in formula (4), which may be the same or different, each represent a hydrogen atom, a halogen atom or a monovalent group, or are bound to each other to form a ring,

$R_a$  in formula (1) represents a group having a site capable of being bound to at least 3 boron atoms which are the same or different,  $R_b$  in formula (2) represents a group having a site capable of being bound to at least 4 boron atoms which are the same or different,  $R_c$  in formula (3) represents a group having a site capable of being bound to at least 5 boron atoms which are the same or different, and  $R_d$  in formula (4) represents a group having a site capable of being bound to at least 6 boron atoms which are the same or different.

3. (Amended) The polymeric electrolyte according to claim 2, [characterized in that] wherein  $R^{11}, R^{12}, R^{13}, R^{14}, R^{15}, R^{16}, R^{21}, R^{22}, R^{23}, R^{24}, R^{25}, R^{26}, R^{27}, R^{28}, R^{31}, R^{32}, R^{33}, R^{34}, R^{35}, R^{36}, R^{37}, R^{38}, R^{39}, R^{310}, R^{41}, R^{42}, R^{43}, R^{44}, R^{45}, R^{46}, R^{47}, R^{48}, R^{49}, R^{410}, R^{411}$  and  $R^{412}$  are one or more [groups selected from the member consisting] of an alkyl group, an aryl group and flourine- substituted derivatives thereof.

4. (Amended) The polymeric electrolyte according to any one of claims 1 to 3, [characterized in that] wherein the polymeric compound is one or more selected from the group consisting of a polyalkylene, a polyether, a polyester, a polyamine, a polyimide, a polyurethane, a polysulfide, a polyphosphazene, a

polysiloxane, derivatives thereof, copolymers thereof and crosslinked products thereof.

5. (Amended) The polymeric electrolyte according to any one of claims 1 to 3, [characterized in that] wherein the polymeric compound is one or more selected from the group consisting of a polyalkylene oxide, polyvinylidene fluoride, polyhexafluoropropylene, polyacrylonitrile, polymethyl methacrylate, derivatives thereof, copolymers thereof and crosslinked products thereof.

6. (Amended) The polymeric electrolyte according to any one of claims 1 to [5] 3, [characterized in that] wherein the electrolytic salt is a metallic salt.

7. (Amended) The polymeric electrolyte according to claim 6, [characterized in that] wherein the metallic salt is a lithium salt.

8. (Amended) The polymeric electrolyte according to claim 7, [characterized in that] wherein the lithium salt is one or more selected from the group consisting of LiBF<sub>4</sub>, LiPF<sub>6</sub>, LiClO<sub>4</sub>, LiAsF<sub>6</sub>, LiCF<sub>3</sub>SO<sub>3</sub>, LiN(CF<sub>3</sub>SO<sub>2</sub>)<sub>2</sub>, LiN(C<sub>2</sub>F<sub>5</sub>SO<sub>2</sub>)<sub>2</sub>, LiC(CF<sub>3</sub>SO<sub>2</sub>)<sub>3</sub>, LiCl, LiF, LiBr, LiI, derivatives thereof and like.

9. (Amended) The polymeric electrolyte according to any one of claims 1 to [8] 3, [characterized by] further [containing] comprising a nonaqueous solvent.

10. (Amended) The polymeric electrolyte according to claim 9, [characterized in that] wherein the nonaqueous solvent is an aprotic solvent.

11. (Amended) The polymeric electrolyte according to claim 10, [characterized in that] wherein the aprotic solvent is one or more selected from the group consisting of carbonates, lactones, ethers, sulfolanes and dioxolanes.

12. (Amended) The polymeric electrolyte according to any one of claims 1 to [11] 3, [characterized in that] wherein the molar ratio of the compound having boron atoms [in the structure] to the electrolytic salt is 0.1:100 to 300:100.

13. (Amended) An electric device [using] comprising the polymeric electrolyte according to any one of claims 1 to [12] 3.

14. (Amended) A cell [in which] comprising a positive electrode [and], a negative electrode [are linked through] and the polymeric electrolyte according to any one of claims 1 to [12] 3, said electrodes being linked through said electrolyte.

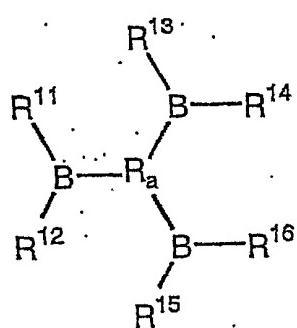
15. (Amended) The cell according to claim 14, [characterized in that] wherein the positive electrode is made of a double metal oxide capable of occluding and releasing lithium ions, and the negative electrode is made of a lithium metal, a lithium alloy or a compound capable of reversibly occluding and releasing lithium ions.

16. (Amended) A nonaqueous electrolyte [made of] comprising an electrolytic salt, [and] a nonaqueous solvent that dissolves the electrolytic salt, [characterized by containing] and a compound having boron atoms [in the structure].

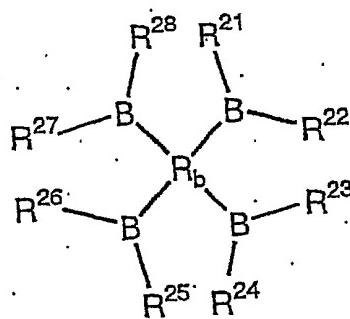
17. (Amended) The nonaqueous electrolyte according to claim 16, [characterized in that] wherein the compound having boron atoms [in the structure]

is one or more selected from the group consisting of compounds represented by the following general formulas (1) to (4)

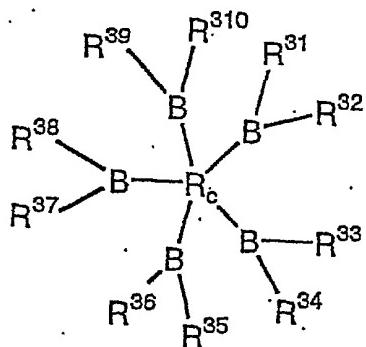
general formula (1)



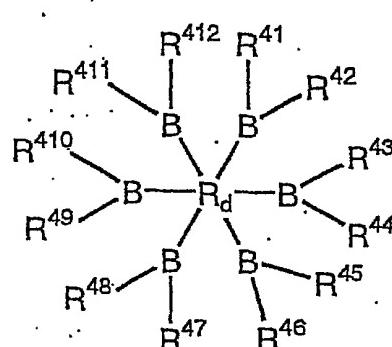
general formula (2)



general formula (3)



general formula (4)



wherein

$R^{11}, R^{12}, R^{13}, R^{14}, R^{15}$  and  $R^{16}$  in formula (1),  $R^{21}, R^{22}, R^{23}, R^{24}, R^{25}, R^{26}, R^{27}$  and  $R^{28}$  in formula (2),  $R^{31}, R^{32}, R^{33}, R^{34}, R^{35}, R^{36}, R^{37}, R^{38}, R^{39}$  and  $R^{310}$  in formula (3), and  $R^{41}, R^{42}, R^{43}, R^{44}, R^{45}, R^{46}, R^{47}, R^{48}, R^{49}, R^{410}, R^{411}$  and  $R^{412}$  in formula (4),

which may be the same or different, each represent a hydrogen atom, a halogen atom or a monovalent group, or are bound to each other to form a ring, Ra in formula (1) represents a group having a site capable of being bound to at least 3 boron atoms which are the same or different, Rb in formula (2) represents a group having a site capable of being bound to at least 4 boron atoms which are the same or different, Rc in formula (3) represents a group having a site capable of being bound to at least 5 boron atoms which are the same or different, and Rd in formula (4) represents a group having a site capable of being bound to at least 6 boron atoms which are the same or different.

18. (Amended) The nonaqueous electrolyte according to claim 17, [characterized in that] wherein R<sup>11</sup>, R<sup>12</sup>, R<sup>13</sup>, R<sup>14</sup>, R<sup>15</sup>, R<sup>16</sup>, R<sup>21</sup>, R<sup>22</sup>, R<sup>23</sup>, R<sup>24</sup>, R<sup>25</sup>, R<sup>26</sup>, R<sup>27</sup>, R<sup>28</sup>, R<sup>31</sup>, R<sup>32</sup>, R<sup>33</sup>, R<sup>34</sup>, R<sup>35</sup>, R<sup>36</sup>, R<sup>37</sup>, R<sup>38</sup>, R<sup>39</sup>, R<sup>310</sup>, R<sup>41</sup>, R<sup>42</sup>, R<sup>43</sup>, R<sup>44</sup>, R<sup>45</sup>, R<sup>46</sup>, R<sup>47</sup>, R<sup>48</sup>, R<sup>49</sup>, R<sup>410</sup>, R<sup>411</sup> and R<sup>412</sup> are one or more [groups selected from the member consisting] of an alkyl group, an aryl group and fluorine-substituted derivatives thereof.

19. (Amended) The nonaqueous electrolyte according to any one of claims 16 to 18, [characterized in that] wherein the electrolytic salt is a metallic salt.

20. (Amended) The nonaqueous electrolyte according to claim 19, [characterized in that] wherein the metallic salt is a lithium salt.

21. (Amended) The nonaqueous electrolyte according to claim 20, [characterized in that] wherein the lithium salt is one or more selected from the group consisting of LiBF<sub>4</sub>, LiPF<sub>6</sub>, LiClO<sub>4</sub>, LiAsF<sub>6</sub>, LiCF<sub>3</sub>SO<sub>3</sub>, LiN(CF<sub>3</sub>SO<sub>2</sub>)<sub>2</sub>, LiN(C<sub>2</sub>F<sub>5</sub>SO<sub>2</sub>)<sub>2</sub>, LiC(CF<sub>3</sub>SO<sub>2</sub>)<sub>3</sub>, LiCl, LiF, LiBr, LiI, derivatives thereof and the like.

22. (Amended) The nonaqueous electrolyte according to any one of claims 16 to [21] 18, characterized in that the nonaqueous solvent is an aprotic solvent.

23. (Amended) The nonaqueous electrolyte according to claim 22, [characterized in that] wherein the aprotic solvent is one or more selected from the group consisting of carbonates, lactones, ethers, sulfolanes and dioxolanes.

24. (Amended) The nonaqueous electrolyte according to any one of claims 16 to [23] 18, [characterized in that] wherein the molar ratio of the compound having the boron atoms in the structure to the electrolytic salt is 0.1:100 to 300:100.

25. (Amended) An electric device [using] comprising the nonaqueous electrolyte according to any one of claims 16 to [24] 18.

26. (Amended) A cell [in which] comprising a positive electrode, [and] a negative electrode, [are linked through] the nonaqueous electrolyte according to any one of claims 16 to [24] 18 and a separator, said electrodes being linked through said electrolytes and said separator.

27. (Amended) The cell according to claim 26, [characterized in that] wherein the positive electrode is made of a double metal oxide capable of occluding and releasing lithium ions, and the negative electrode is made of a lithium metal, a lithium alloy or a compound capable of reversibly occluding and releasing lithium ions.

DESCRIPTION

**POLYMERIC ELECTROLYTE, NONAQUEOUS ELECTROLYTE AND  
ELECTRIC DEVICE USING THE SAME**

**Technical Field**

The present invention relates to a polymeric electrolyte and a nonaqueous electrolyte which can improve a transport rate of charge carrier ions upon using a compound having boron atoms in the structure as an additive, and an electric device using the same.

**Background of the Invention**

According to the development of cells having a high voltage and a high capacity, a large number of various polymeric electrolytes and nonaqueous electrolytes have been proposed. However, polymeric electrolytes and nonaqueous electrolytes have an ionic conductivity which is lower than that of aqueous electrolytes by more than one figure. Further, for example, a polymeric electrolyte using polyethylene glycol has defects that it is low in transfer and transport rates of charge carrier ions. Thus, attempts of improvement have been made by using various methods.

Meanwhile, when nonaqueous electrolytes are applied to cells, they are problematic in that charge/discharge

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efficiency and cycle characteristics are low. Therefore, in order to improve these, a solvent composition of an electrolytic solution and a type of a support salt has been studied, and a system in which an additive is added to a nonaqueous electrolyte has been studied. For example, Japanese Patent Laid-Open No. 3,728/1999 discloses a nonaqueous electrolytic solution secondary cell comprising a positive electrode and a negative electrode containing a material capable of occluding and releasing lithium reversibly, a nonaqueous electrolytic solution containing a lithium salt and a separator, at least one organic boron compound being contained in the cell in a predetermined amount.

However, these prior techniques have posed a problem that an effective concentration region of an additive is narrow and further an effect provided by its addition per weight is small.

In view of the foregoing, the invention has been made, and aims to provide a polymeric electrolyte and a nonaqueous electrolyte which increases a dissociation degree of an electrolytic salt, which can improve a transport rate of charge carrier ions and in which an effective concentration region of an additive is wide and an effect provided by its addition per weight is great, and an electric device using the same.

### **Disclosure of the Invention**

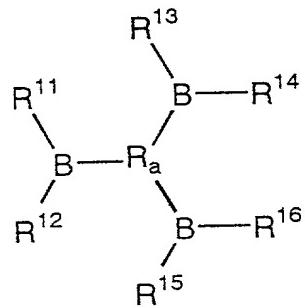
The present inventors have assiduously conducted investigations to solve the problems. They have consequently conceived that counter ions of charge carrier ions are trapped and less moved to control a transport rate of charge carrier ions, and have found that the use of a compound containing plural trivalent boron atoms in the structure, a Lewis acid, as an additive, is effective for solving the problems. This finding has led to the completion of the invention.

That is, the polymeric electrolyte of the invention is a polymeric electrolyte made of an electrolytic salt and a polymeric compound forming a complex with the electrolytic salt, the polymeric electrolyte containing one or more compounds having boron atoms in the structure.

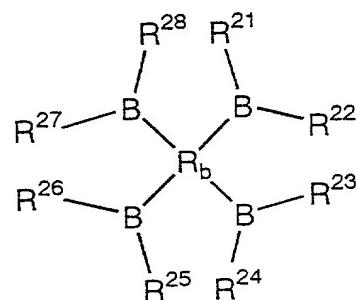
Further, the nonaqueous electrolyte of the invention is a nonaqueous electrolyte made of an electrolytic salt and a nonaqueous solvent that dissolves the electrolytic salt, the nonaqueous electrolyte containing one or more compounds having boron atoms in the structure.

The compound having boron atoms in the structure is preferably selected from the group consisting of compounds represented by the following general formulas (1) to (4).

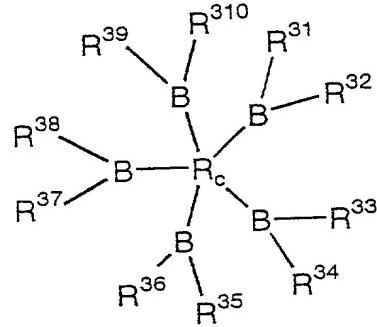
general formula (1)



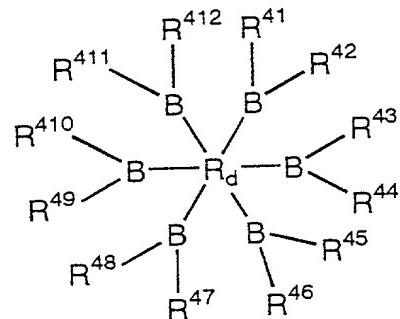
general formula (2)



general formula (3)



general formula (4)



R<sup>11</sup>, R<sup>12</sup>, R<sup>13</sup>, R<sup>14</sup>, R<sup>15</sup> and R<sup>16</sup> in formula (1), R<sup>21</sup>, R<sup>22</sup>, R<sup>23</sup>, R<sup>24</sup>, R<sup>25</sup>, R<sup>26</sup>, R<sup>27</sup> and R<sup>28</sup> in formula (2), R<sup>31</sup>, R<sup>32</sup>, R<sup>33</sup>, R<sup>34</sup>, R<sup>35</sup>, R<sup>36</sup>, R<sup>37</sup>, R<sup>38</sup>, R<sup>39</sup> and R<sup>310</sup> in formula (3), and R<sup>41</sup>, R<sup>42</sup>, R<sup>43</sup>, R<sup>44</sup>, R<sup>45</sup>, R<sup>46</sup>, R<sup>47</sup>, R<sup>48</sup>, R<sup>49</sup>, R<sup>410</sup>, R<sup>411</sup> and R<sup>412</sup> in formula (4), which may be the same or different, each represent a hydrogen atom, a halogen atom or a monovalent group, or are bound to each other to form a ring.

R<sub>a</sub> in formula (1) represents a group having a site capable of being bound to at least 3 boron atoms which are the same or different, R<sub>b</sub> in formula (2) represents a group having a site capable of being bound to at least 4 boron atoms

which are the same or different,  $R_c$  in formula (3) represents a group having a site capable of being bound to at least 5 boron atoms which are the same or different, and  $R_d$  in formula (4) represents a group having a site capable of being bound to at least 6 boron atoms which are the same or different.

The polymeric compound used in the polymeric electrolyte of the invention includes, for example, a polyalkylene, a polyether, a polyester, a polyamine, a polyimide, a polyurethane, a polysulfide, a polyphosphazene, a polysiloxane, derivatives thereof, copolymers thereof and crosslinked products thereof. Alternatively, it includes a polyalkylene oxide, polyvinylidene fluoride, polyhexafluoropropylene, polyacrylonitrile, polymethyl methacrylate, derivatives thereof, copolymers thereof and crosslinked products thereof.

As the electrolytic salt, a lithium salt is preferably used. Further, as the nonaqueous solvent, an aprotic solvent is preferably used.

The electric device of the invention is manufactured using any of the polymeric electrolytes and the nonaqueous electrolytes. For example, when the electric device is a cell, it is obtained by linking a positive electrode and a negative electrode through any of the polymeric electrolytes or through any of the nonaqueous electrolytes and a separator.

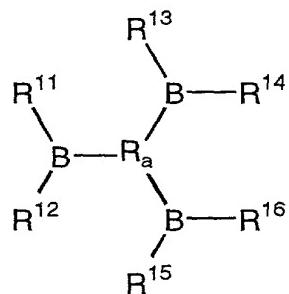
## **Best Mode For Carrying Out the Invention**

Preferable embodiments of the invention are described below. However, the invention is not limited thereto.

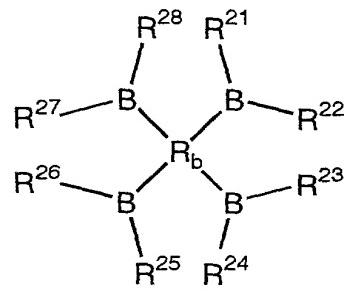
### **1. Compound having boron atoms**

With respect to the compound having boron atoms in the structure, which is added to a polymeric electrolyte or a nonaqueous electrolyte of the invention, the compounds represented by general formulas (1) to (4) are preferably used as described earlier.

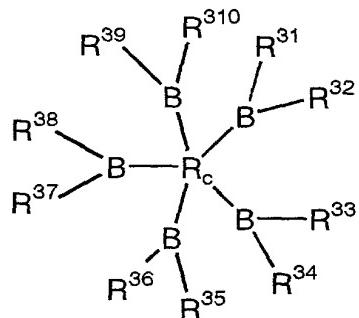
general formula (1)



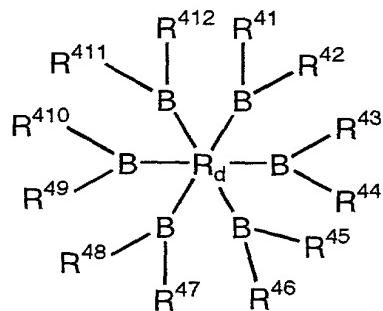
general formula (2)



general formula (3)



general formula (4)



R<sup>11</sup>, R<sup>12</sup>, R<sup>13</sup>, R<sup>14</sup>, R<sup>15</sup> and R<sup>16</sup> in formula (1), R<sup>21</sup>, R<sup>22</sup>, R<sup>23</sup>, R<sup>24</sup>, R<sup>25</sup>, R<sup>26</sup>, R<sup>27</sup> and R<sup>28</sup> in formula (2), R<sup>31</sup>, R<sup>32</sup>, R<sup>33</sup>, R<sup>34</sup>, R<sup>35</sup>,

$R^{36}$ ,  $R^{37}$ ,  $R^{38}$ ,  $R^{39}$  and  $R^{310}$  in formula (3), and  $R^{41}$ ,  $R^{42}$ ,  $R^{43}$ ,  $R^{44}$ ,  $R^{45}$ ,  $R^{46}$ ,  $R^{47}$ ,  $R^{48}$ ,  $R^{49}$ ,  $R^{410}$ ,  $R^{411}$  and  $R^{412}$  in formula (4), which may be the same or different, each represent a hydrogen atom, a halogen atom or a monovalent group.

Examples of the monovalent group include an alkyl group, an alkoxy group, an aryl group, an alkenyl group, an alkinyl group, an aralkyl group, a cycloalkyl group, a cyano group, a hydroxyl group, a formyl group, an aryloxy group, an alkylthio group, an arylthio group, an acyloxy group, a sulfonyloxy group, an amino group, an alkylamino group, an arylamino group, a carboxyamino group, an oxysulfonylamino group, a sulfonamido group, an oxycarbonylamino group, a ureido group, an acyl group, an oxycarbonyl group, a carbamoyl group, a sulfonyl group, a sulfinyl group, an oxysulfonyl group, a sulfamoyl group, a carboxylic group, a sulfonic group, a phosphonic group, a heterocyclic group,  $-B(R^1)(R^2)$ ,  $-OB(R^1)(R^2)$  and  $-OSi(R^1)(R^2)(R^3)$ . Of these, an alkyl group, an aryl group and fluorine-substituted derivatives thereof are preferable.  $R^1$ ,  $R^2$  and  $R^3$  herein each represent a hydrogen atom, a halogen atom or a monovalent group. Examples of the monovalent group include an alkyl group, an alkoxy group, an aryl group, an alkenyl group, an alkinyl group, an aralkyl group, a cycloalkyl group, a cyano group, a hydroxyl group, a formyl group, an aryloxy group, an alkylthio group, an arylthio group, an acyloxy group, a sulfonyloxy group, an

amino group, an alkylamino group, an arylamino group, a carboxyamino group, an oxysulfonylamino group, a sulfonamide group, an oxycarbonylamino group, a ureido group, an acyl group, an oxycarbonyl group, a carbamoyl group, a sulfonyl group, a sulfinyl group, an oxysulfonyl group, a sulfamoyl group, a carboxylic group, a sulfonic group, a phosphonic group, a heterocyclic group and derivatives thereof.

Further,  $R^{11}$ ,  $R^{12}$ ,  $R^{13}$ ,  $R^{14}$ ,  $R^{15}$  and  $R^{16}$  in formula (1),  $R^{21}$ ,  $R^{22}$ ,  $R^{23}$ ,  $R^{24}$ ,  $R^{25}$ ,  $R^{26}$ ,  $R^{27}$  and  $R^{28}$  in formula (2),  $R^{31}$ ,  $R^{32}$ ,  $R^{33}$ ,  $R^{34}$ ,  $R^{35}$ ,  $R^{36}$ ,  $R^{37}$ ,  $R^{38}$  and  $R^{310}$  in formula (3), and  $R^{41}$ ,  $R^{42}$ ,  $R^{43}$ ,  $R^{44}$ ,  $R^{45}$ ,  $R^{46}$ ,  $R^{47}$ ,  $R^{48}$ ,  $R^{49}$ ,  $R^{410}$ ,  $R^{411}$  and  $R^{412}$  in formula (4) may be bound to each other to form a ring, and this ring may have a substituent. Moreover, each group may be substituted with a group which can be substituted.

Examples of the group having a site capable of being bound to at least 3 boron atoms which are the same or different, as represented by Ra, in formula (1) include residues such as glycerin, trimethylolethane, trihydroxybenzene, dihydroxybenzoic acid, diaminobenzoic acid, tribromobenzene and the like.

Examples of the group having a site capable of being bound to at least 4 boron atoms which are the same or different, as represented by Rb, in formula (2) include residues such as diglycerin, pentaerythritol, tetrabromobenzene and the like.

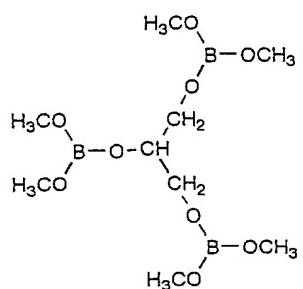
Examples of the group having a site capable of being bound to at least 5 boron atoms which are the same or different, as represented by  $R_c$ , in formula (3) include residues such as glucose, morin and the like.

Examples of the group having a site capable of being bound to at least 6 boron atoms which are the same or different, as represented by  $R_d$ , in formula (4) include residues such as galactaric acid, myricetin and the like.

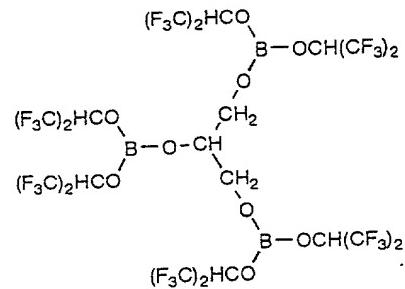
The compounds represented by the foregoing general formulas (1) to (4) may be used either singly or in combination.

Specific examples of the compounds represented by general formulas (1) to (4) are as follows.

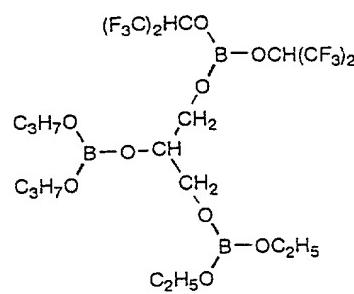
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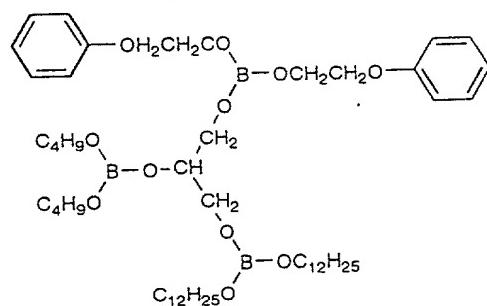
3Z-2



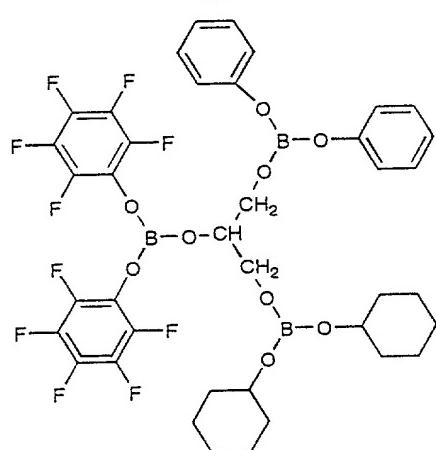
3Z-3



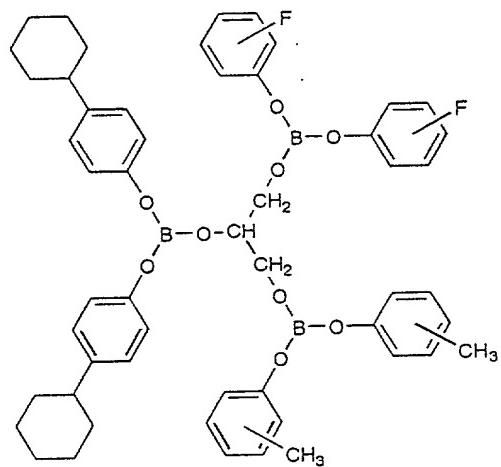
3Z-4



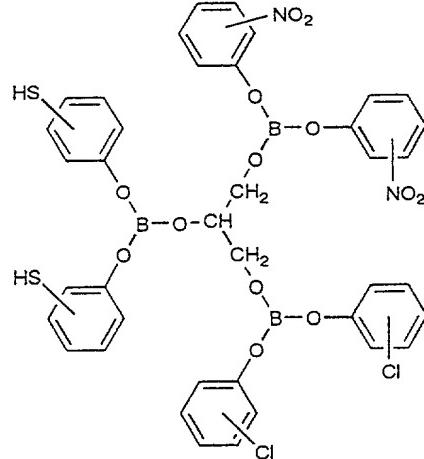
3Z-5



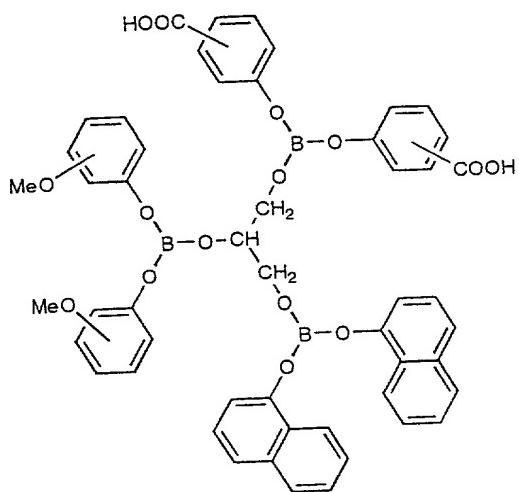
3Z-6



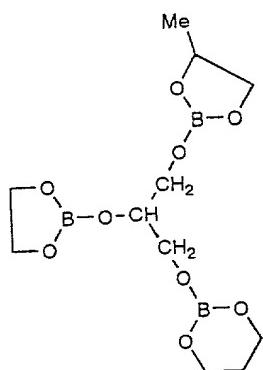
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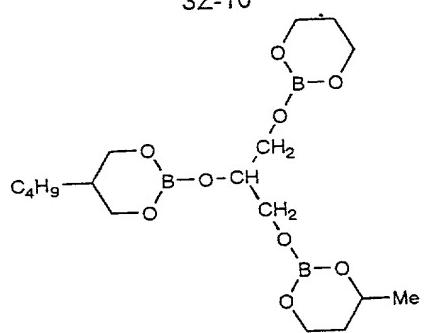
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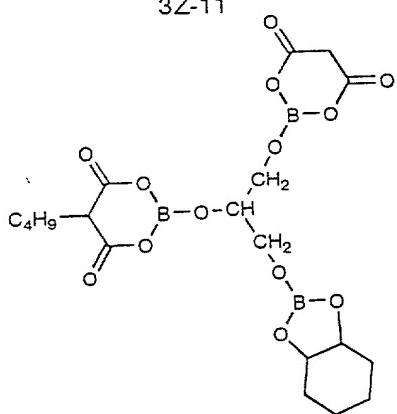
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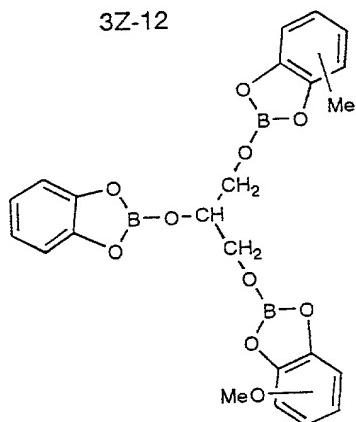
3Z-10



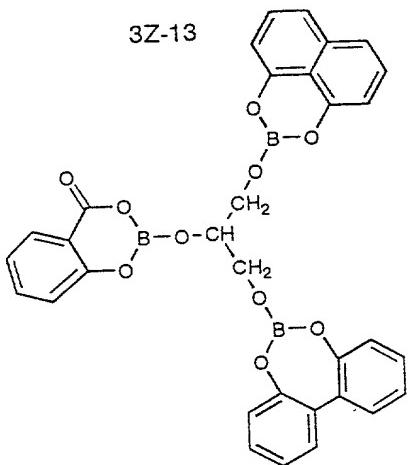
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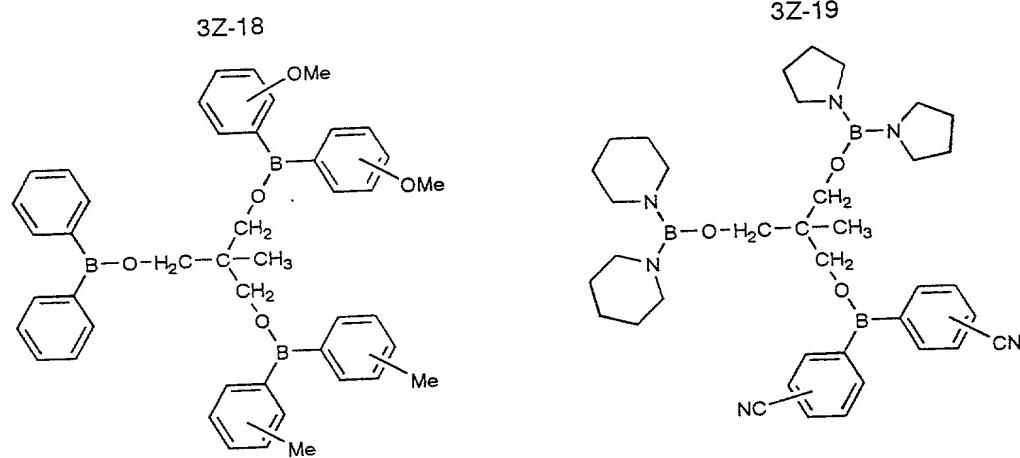
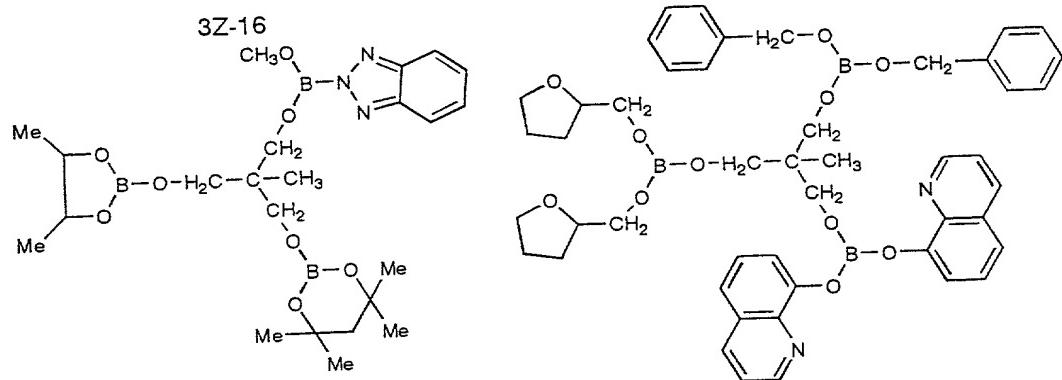
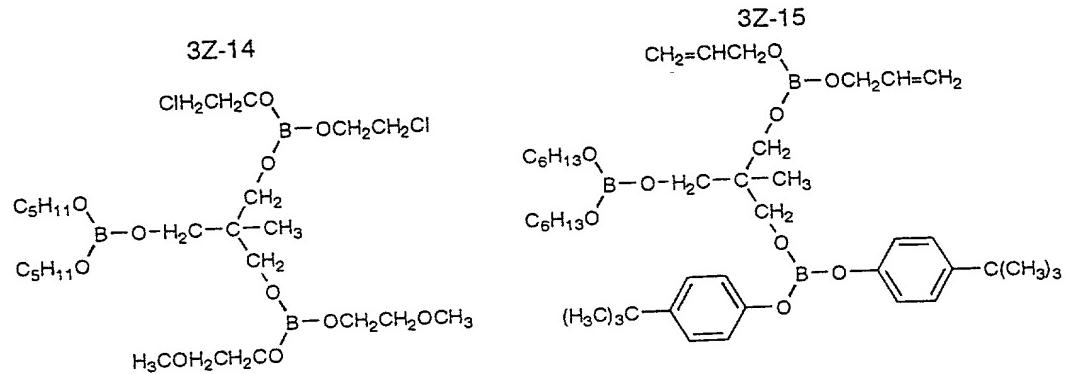


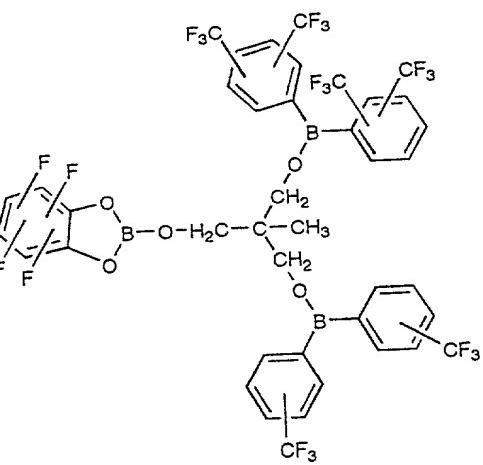
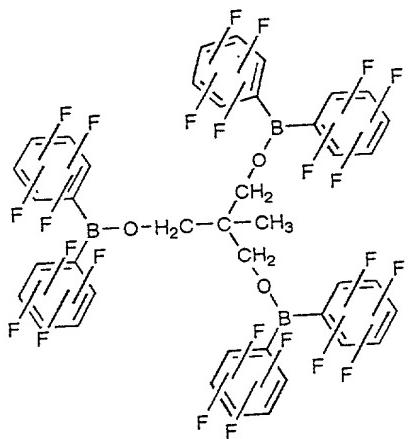
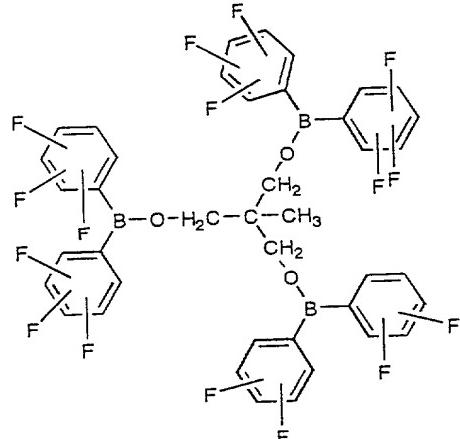
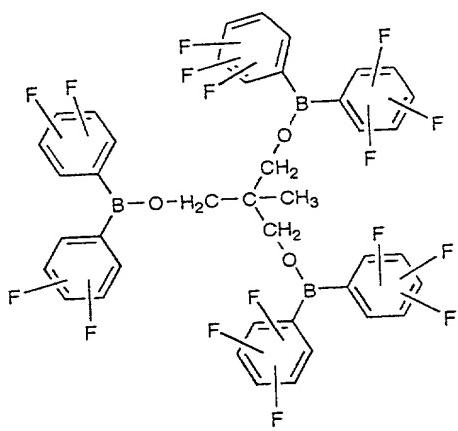
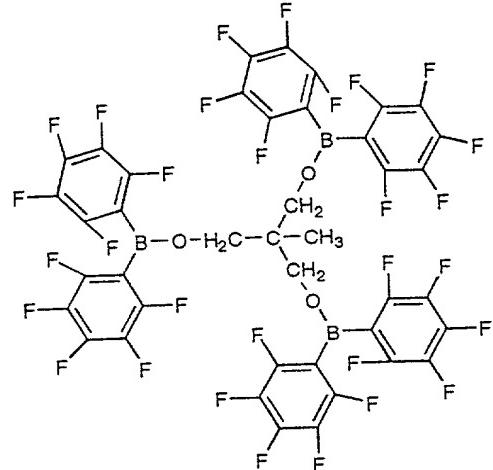
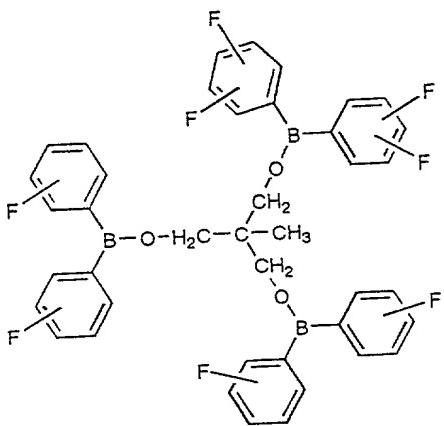
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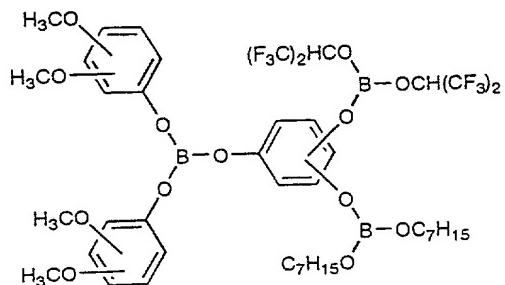
3Z-13



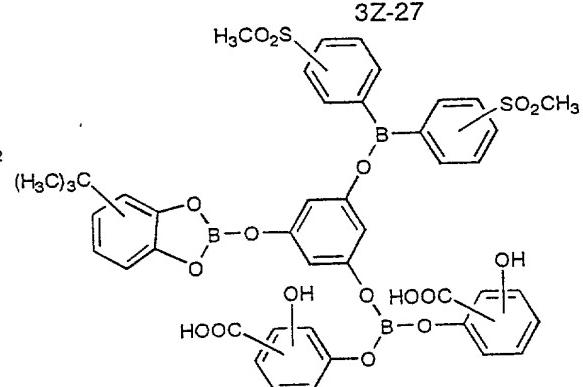




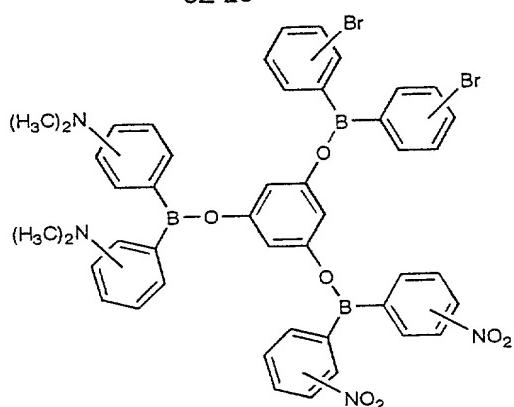
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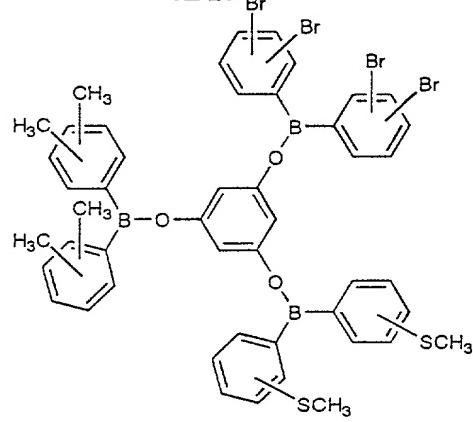
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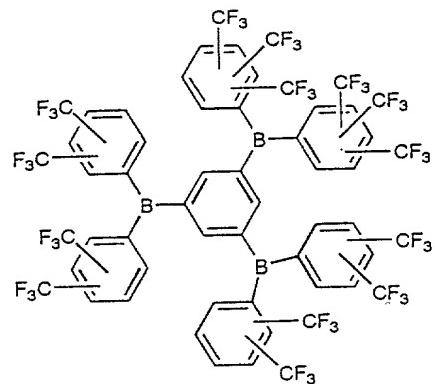
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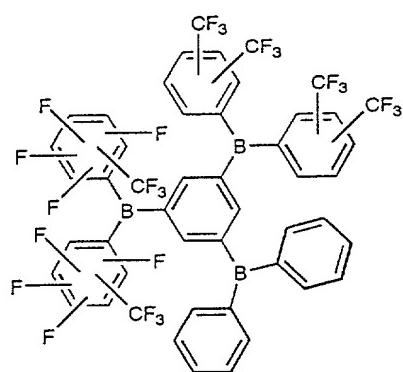
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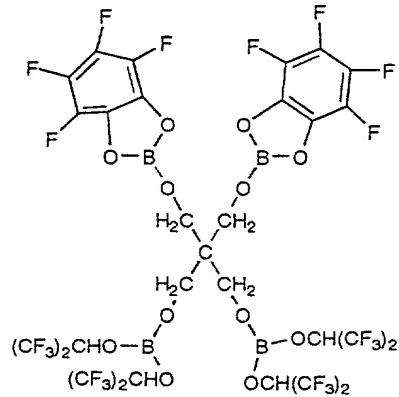
3Z-30



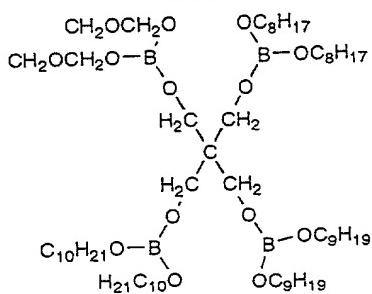
3Z-31



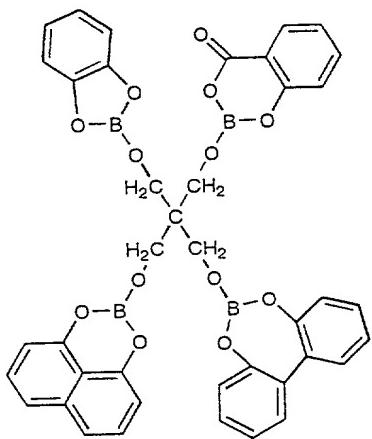
4Z-3



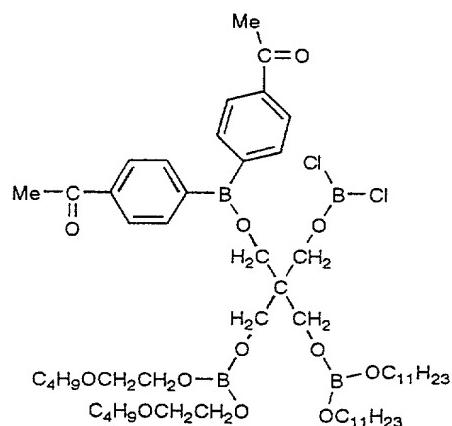
4Z-1



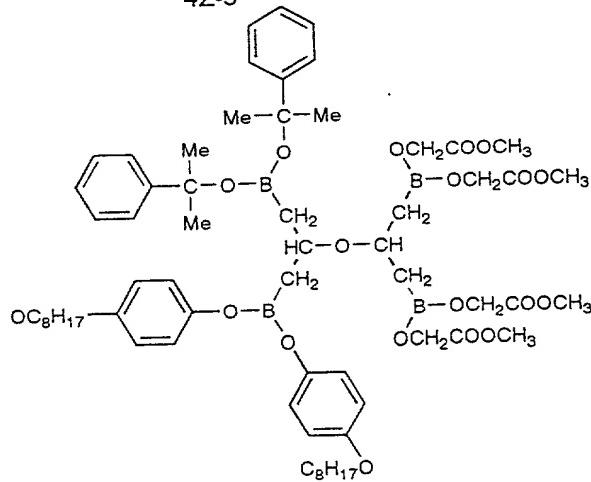
4Z-2



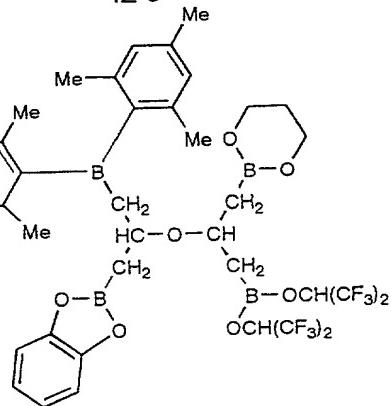
4Z-4



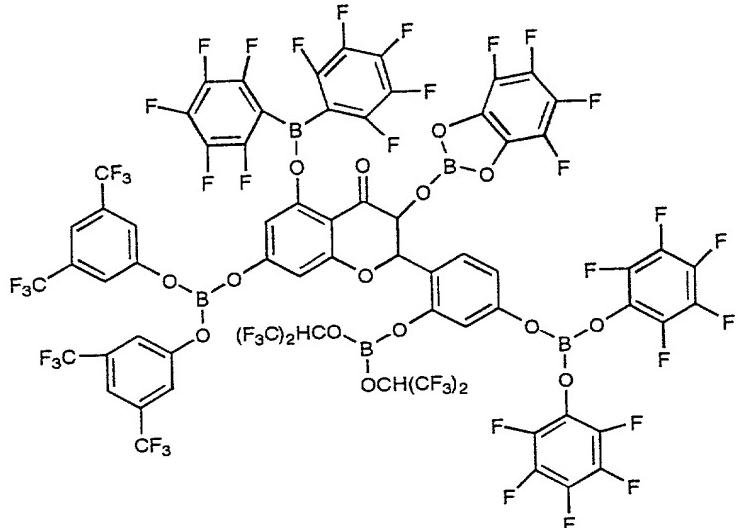
4Z-5



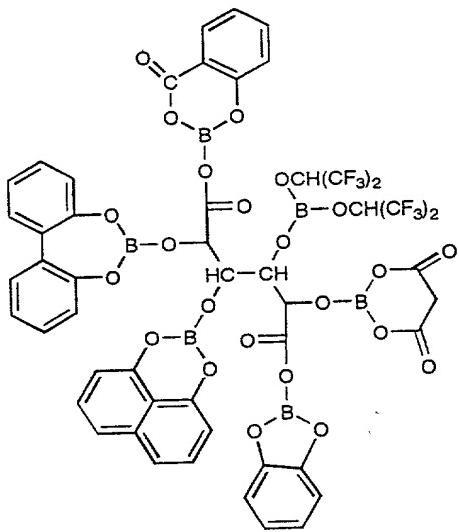
4Z-6



5Z-1



6Z-1

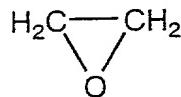


## 2. Polymeric electrolyte

Examples of the polymeric compound used as the polymeric electrolyte of the invention include a polyalkylene, a polyether, a polyester, a polyamine, a polyimide, a polyurethane, a polysulfide, a polyphosphazene, a polysiloxane, a polyalkylene oxide, polyvinylidene fluoride, polyhexafluoropropylene, polyacrylonitrile, polymethyl methacrylate, derivatives thereof, copolymers thereof and crosslinked products thereof. They are used either singly or in combination.

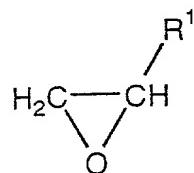
Of these, a polymeric compound made of a polyalkylene oxide, and/or derivatives thereof, and/or crosslinked products thereof is preferable. For example, a polymer or a copolymer of compound (A) represented by the following formula (5) and/or compound (B) represented by the following formula (6) can be used.

compound (A)



(5)

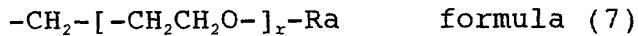
compound (B)



(6)

In formula (6), R<sup>1</sup> represents a methyl group, an ethyl

group, a propyl group, a butyl group or a group represented by the following formula (7).



In formula (7), r represents 0 or an integer of 1 or more, and Ra represents a methyl group, an ethyl group, a propyl group or a butyl group.

The electrolytic salt is preferably a metallic salt, more preferably a lithium salt. Examples thereof include LiBF<sub>4</sub>, LiPF<sub>6</sub>, LiClO<sub>4</sub>, LiAsF<sub>6</sub>, LiCF<sub>3</sub>SO<sub>3</sub>, LiN(CF<sub>3</sub>SO<sub>2</sub>)<sub>2</sub>, LiN(C<sub>2</sub>F<sub>5</sub>SO<sub>2</sub>)<sub>2</sub>, LiC(CF<sub>3</sub>SO<sub>2</sub>)<sub>3</sub>, LiCl, LiF, LiBr, LiI, derivatives thereof and like. These lithium salts may be used either singly or in combination.

The concentration of these electrolytic salts is usually 0.01 mol/kg to 10 mols/kg, preferably 0.2 mol/kg to 6 mols/kg.

The nonaqueous solvent is preferably an aprotic solvent. Examples thereof include carbonates, lactones, ethers, sulfolanes and dioxolanes. These aprotic solvents may be used either singly or in combination.

The polymeric compound to aprotic solvent mixing ratio is 1/99 to 99/1, preferably 5/95 to 95/5, more preferably 10/90 to 90/10 in terms of a weight ratio.

Further, a molar ratio of the compound having boron atoms in the structure to the electrolytic salt is preferably 0.1:100 to 300:100, more preferably 1:100 to 200:100, further

preferably 50:100 to 100:100.

### **3. Nonaqueous electrolyte**

Examples of the electrolytic salt and the nonaqueous solvent used in the nonaqueous electrolyte of the invention are the same as those listed on the foregoing polymeric electrolyte. Further, the concentration of the electrolytic salt and the molar ratio of the boron atom-containing compound to the electrolytic salt may be in the same ranges as mentioned on the foregoing polymeric electrolyte.

### **4. Electric devices**

The polymeric electrolyte of the invention can be applied to various electric devices, and examples thereof include cells, capacitors and the like. Typical of these are cells which are obtained by linking a positive electrode and a negative electrode through any of the foregoing polymeric electrolytes or through any of the foregoing nonaqueous electrolytes and a separator.

In the positive electrode herein, a double metal oxide capable of occluding and releasing lithium ions is used. Examples thereof include cobalt lithium oxide, nickel lithium oxide, manganese lithium oxide, vanadium pentoxide and the like.

Further, in the negative electrode, a lithium metal, a lithium alloy or a substance capable of reversibly occluding and releasing lithium ions is used. Examples of such a

substance include carbon and the like.

Moreover, as the separator, a porous film, a nonwoven fabric, a woven fabric or the like which can permeate lithium ions is used. Examples of the material thereof include olefinic polymers such as polyethylene, polypropylene and the like, fluoropolymers, cellulose-based polymers, glass fibers and the like.

## 5. Examples

The invention is illustrated more specifically below by referring to Examples. However, the invention is not limited to these Examples.

### (1) Examples and Comparative Examples on a polymeric electrolyte

#### [Production of monomers]

##### • Monomer A

Potassium hydroxide (0.01 mol) was added to 1 mol of ethylene glycol monobutyl ether as a starting material, and a vessel was purged with nitrogen while stirring the mixture. Subsequently, the pressure inside the vessel was reduced using a vacuum pump. The temperature was then raised to 120°C, and the reaction was conducted using 1 mol of ethylene oxide as a monomer. After the completion of the reaction, the reaction mixture was cooled until the temperature inside the vessel reached room temperature. A methanol solution of 1.1 mols of sodium methylate was added, and the temperature was slowly

raised to 50°C while reducing the pressure. After methanol was completely removed, 1.2 mols of epichlorohydrin was added, and the mixture was reacted for 4 hours. After the completion of the reaction, adsorption treatment was conducted. After dehydration under reduced pressure, the residue was filtered to obtain a desired product.

• Monomer B

A desired product was obtained in the same manner as monomer A except that ethylene glycol monomethyl ether was used as a starting material and 9 mols of ethylene oxide was used as a monomer.

• Monomer C

A desired product was obtained in the same manner as monomer A except that ethylene glycol monopropyl ether was used as a starting material and 2 mols of ethylene oxide was used as a monomer.

• Monomer D

A desired product was obtained in the same manner as monomer A except that ethylene glycol monoethyl ether was used as a starting material and 49 mols of ethylene oxide was used as a monomer.

• Monomer E

A desired product was obtained in the same manner as monomer A except that ethylene glycol monomethyl ether was used as a starting material and 1 mol of ethylene oxide was

used as a monomer.

[Production of polymeric compound precursors B-1 to B-10]

• Polymeric compound precursor B-1

Potassium hydroxide (0.01 mol) was added to 0.5 mol of ethylene glycol as a starting material, and a vessel was purged with nitrogen while stirring the mixture. The pressure inside the vessel was reduced using a vacuum pump. The temperature was then raised to 120°C, and the reaction was conducted using 38,000 mols of ethylene oxide as a monomer. After the completion of the reaction, the reaction mixture was cooled until the temperature inside the vessel reached room temperature. A methanol solution of 1.1 mols of sodium methylate was added, and the temperature was slowly raised to 50°C while reducing the pressure. After methanol was completely removed, the residue was allowed to cool. One kilogram of toluene was added, and 1 mol of acrylic acid chloride was added. The mixture was reacted for 4 hours. After acid and alkali adsorption treatment was conducted, the residue was filtered, and toluene was removed under reduced pressure to obtain a desired product.

• Polymeric compound precursor B-2

A desired product was obtained in the same manner as polymeric compound precursor B-1 except that 0.33 mol of glycerin was used as a starting material, 28,000 mols of propylene oxide was used as a monomer and methacrylic acid

chloride was used instead of acrylic acid chloride.

· **Polymeric compound precursor B-3**

A desired product was obtained in the same manner as polymeric compound precursor B-1 except that 0.25 mol of diglycerin was used as a starting material, 150 mols of ethylene oxide and 600 mols of 1,2-epoxyhexane were used as monomers, allyl chloride was used instead of acrylic acid chloride.

· **Polymeric compound precursor B-4**

A desired product was obtained in the same manner as polymeric compound precursor B-1 except that 0.5 mol of ethylene glycol was used as a starting material, 2 mols of ethylene oxide and 1 mol of butylene oxide were used as monomers and vinyl chloride was used instead of acrylic acid chloride.

· **Polymeric compound precursor B-5**

A desired product was obtained in the same manner as polymeric compound precursor B-1 except that 0.33 mol of glycerin was used as a starting material and 150 mols of ethylene oxide and 29 mols of 1,2-epoxypentane were used as monomers.

· **Polymeric compound precursor B-6**

A desired product was obtained in the same manner as polymeric compound precursor B-1 except that 600 mols of monomer A was used as a monomer.

· **Polymeric compound precursor B-7**

A desired product was obtained in the same manner as polymeric compound precursor B-2 except that 50 mols of ethylene oxide and 15 mols of monomer B were used as monomers.

**Polymeric compound precursor B-8**

A desired product was obtained in the same manner as polymeric compound precursor B-3 except that 1 mol of ethylene oxide and 1 mol of monomer C were used as monomers.

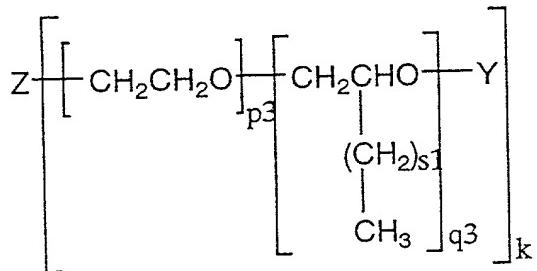
**Polymeric compound precursor B-9**

A desired product was obtained in the same manner as polymeric compound precursor B-4 except that 1,600 mols of ethylene oxide and 400 mols of monomer D were used as monomers.

**Polymeric compound precursor B-10**

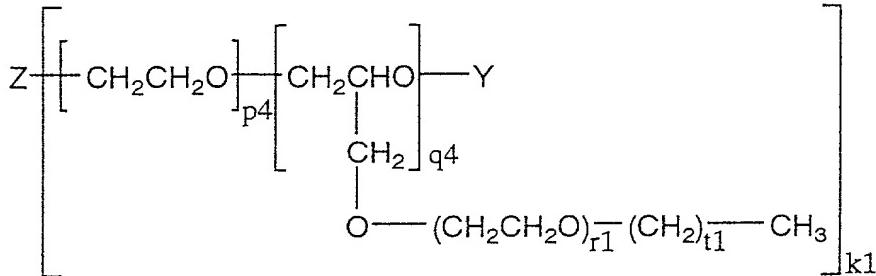
A desired product was obtained in the same manner as polymeric compound precursor B-5 except that 126 mols of ethylene oxide and 24 mols of monomer E were used as monomers.

The structures of polymeric compound precursors B-1 to B-10 obtained in the foregoing production examples are as shown in the following tables and chemical formulas.



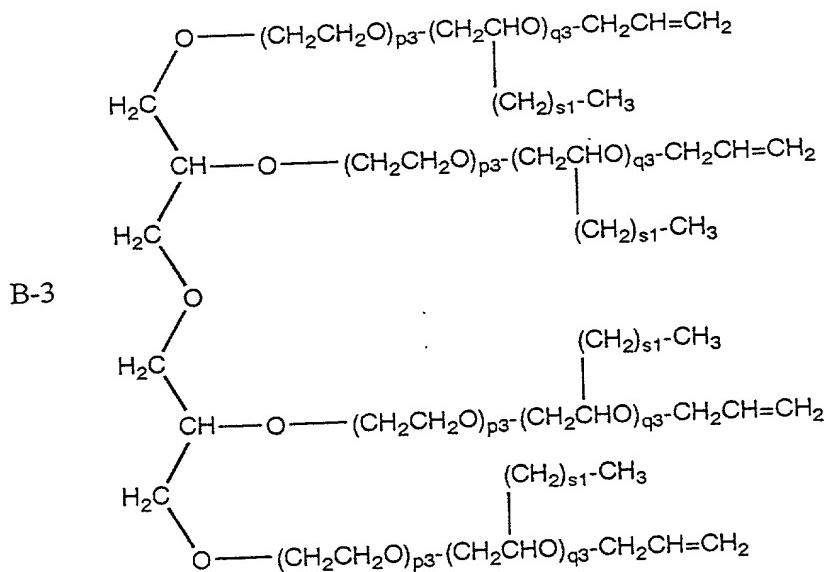
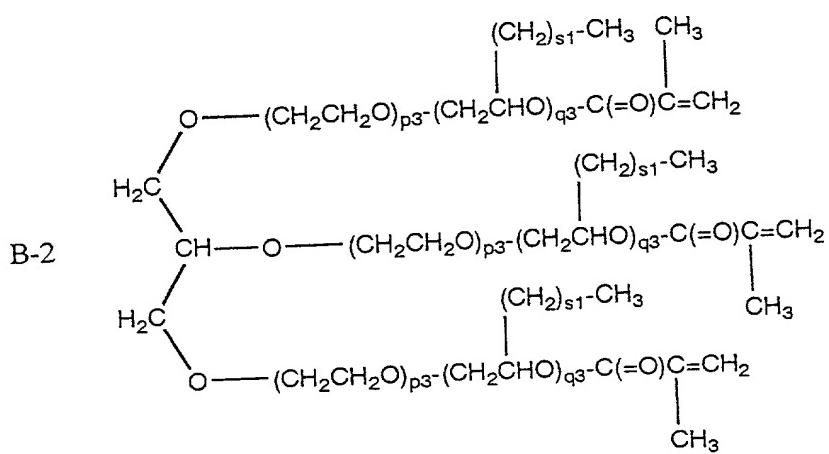
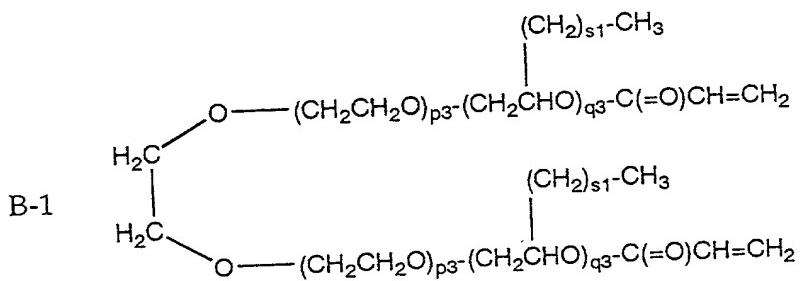
*Z* is an active hydrogen residue.

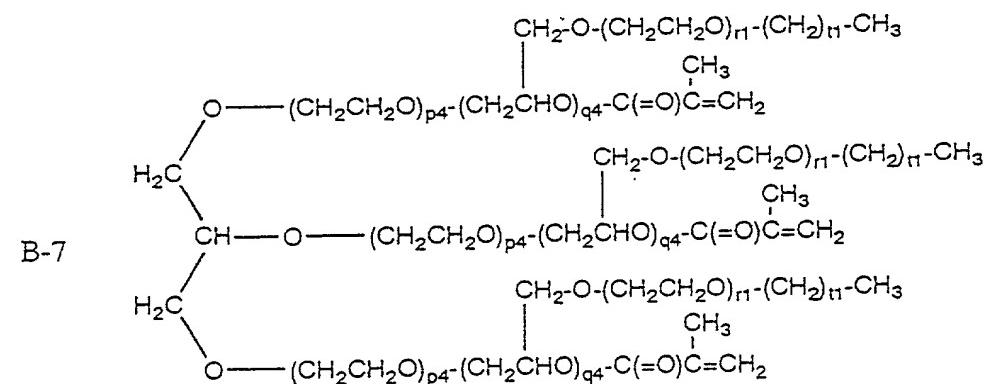
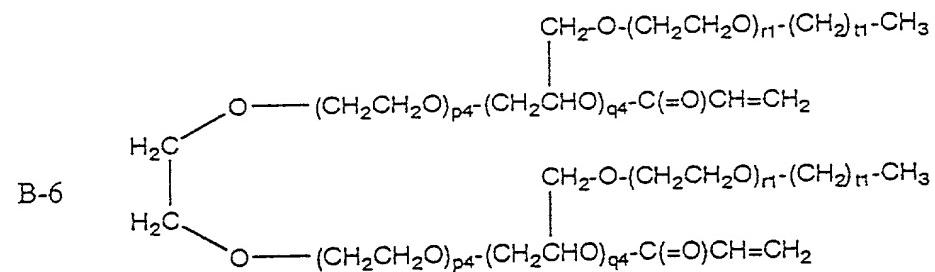
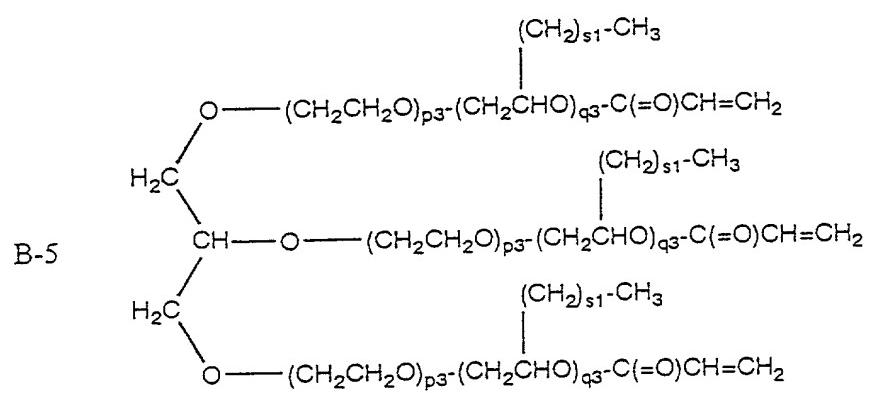
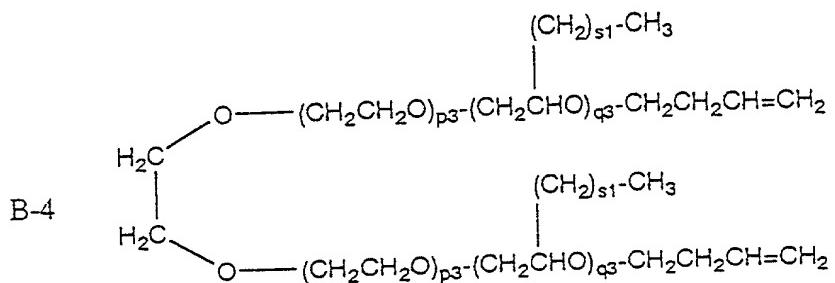
Compound	p <sub>3</sub>	q <sub>3</sub>	s <sub>1</sub>	Y	k
B-1	38000	0	0	acryloyl group	2
B-2	0	28000	1	methacryloyl group	3
B-3	150	600	3	allyl group	4
B-4	2	1	1	vinyl group	2
B-5	150	29	0	acryloyl group	3

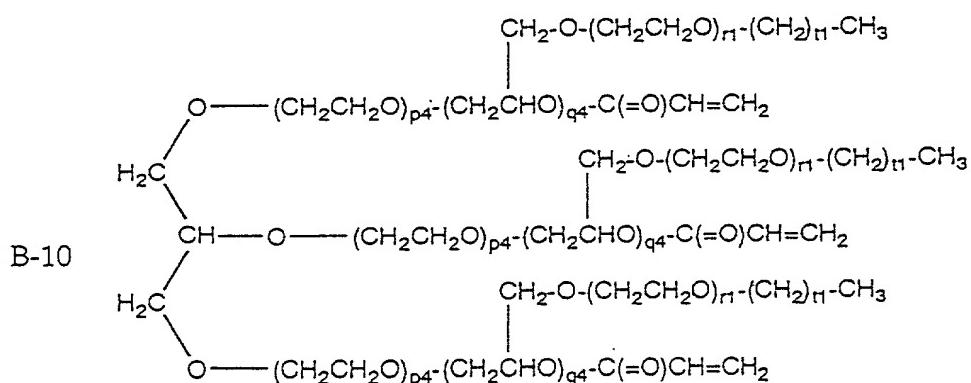
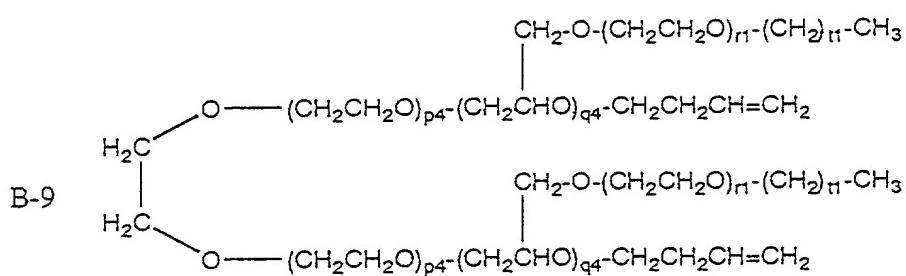
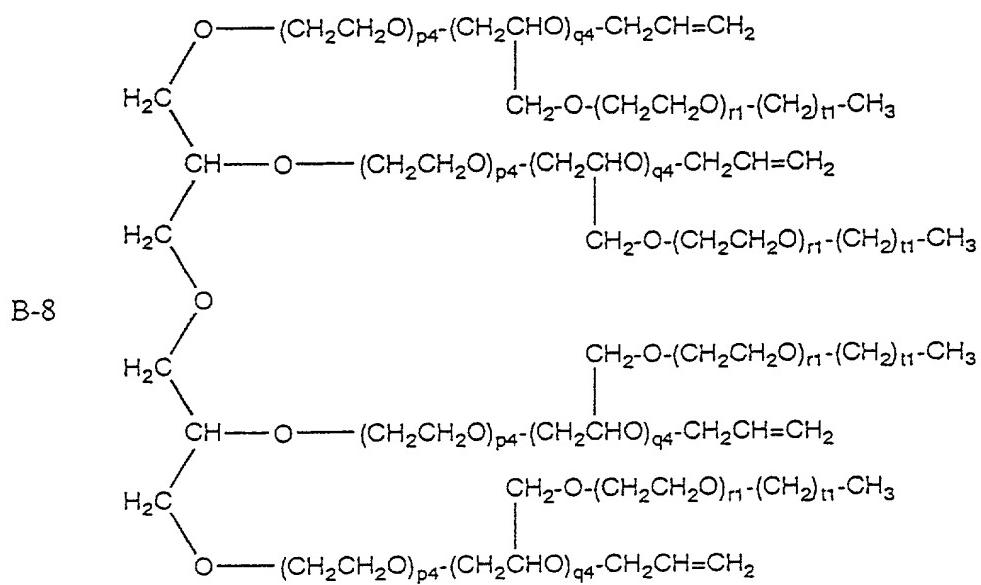


*Z* is an active hydrogen residue.

Compound	p <sub>4</sub>	q <sub>4</sub>	r <sub>1</sub>	t	Y	k
B-6	0	600	2	3	acryloyl group	2
B-7	50	15	10	0	methacryloyl group	3
B-8	1	1	3	2	allyl group	4
B-9	1600	400	50	1	vinyl group	2
B-10	126	24	2	0	acryloyl group	3







[Production of a polymeric electrolyte]

· Example 1

The additive (1 mol/kg) represented by the formula 3Z-3, 1 mol/kg of LiBF<sub>4</sub>, 1.2 g of  $\gamma$ -butyrolactone and a photopolymerization initiator were added to 1 g of polymeric compound precursor B-1. These were dissolved at 40°C, and poured between glass plates. The mixture was then irradiated with ultraviolet rays in an argon atmosphere to obtain a polymeric electrolyte having a thickness of 500  $\mu\text{m}$ .

· Examples 2 to 12

Polymeric electrolytes were obtained in the same manner as in Example 1 except that types and amounts of polymeric compound precursors, additives, salts and aprotic solvents shown in Table 1 were used.

· Comparative Examples 1 to 3

Polymeric electrolytes were obtained in the same manner as in Example 1 except that types and amounts of polymeric compound precursors, additives, salts and aprotic solvents shown in Table 1 were used.

[Measurement of a lithium ion transport rate]

Each of the polymeric electrolytes obtained in the foregoing Examples and Comparative Examples was cut in a circle having a diameter of 13 mm, and this was held between lithium metal electrodes having the same diameter. A lithium ion transport rate was measured by a DC polarization method.

The results are also shown in Table 1.

Table 1

Example	Additive and its concentration (mol/kg)	Polymeric compound precursor	Salt and its concentration (mol/kg)	Aprotic solvent	Lithium ion transport rate
1	3Z-3	1	B-1	LiBF <sub>4</sub>	1 GBL 1.2g 0.84
2	3Z-13	0.03	B-2	LiPF <sub>6</sub>	0.01 - 0.75
3	4Z-3	0.05	B-3	LiClO <sub>4</sub>	0.1 - 0.84
4	5Z-1	0.01	B-4	LiAsF <sub>6</sub>	10 - 0.76
5	6Z-1	0.005	B-5	LiCF <sub>3</sub> SO <sub>3</sub>	5 - 0.83
6	3Z-3	0.05	B-6	LiN(CF <sub>3</sub> SO <sub>2</sub> ) <sub>2</sub>	0.05 - 0.81
7	3Z-13	2	B-7	LiN(C <sub>2</sub> F <sub>5</sub> SO <sub>2</sub> ) <sub>2</sub>	2 - 0.79
8	4Z-3	8	B-8	LiC(CF <sub>3</sub> SO <sub>2</sub> ) <sub>3</sub>	8 - 0.79
9	5Z-1	0.005	B-9	LiCl	0.5 - 0.77
10	6Z-1	0.03	B-10	LiF	3 EC 9.5g, DO 9.5g 0.82
11	3Z-3	0.3	B-5	LiBr	0.3 EC 0.3g, SL 0.3g 0.80
12	3Z-13	6	B-10	LiI	6 EC 1g, DME 2g 0.82
Comparative Example					
1	4Z-3	0.001	B-5	LiN(CF <sub>3</sub> SO <sub>2</sub> ) <sub>2</sub>	0.001 - 0.61
2	5Z-1	15	B-10	LiCl	15 - unmeasurable
3	not added	B-5	1	LiBF <sub>4</sub>	1 - 0.12

\* EC: ethylene carbonate, GBL:  $\gamma$ -butyrolactone, DO: 1,3-dioxolane, DME: 1,2-dimethoxyethane, SL:sulfolane

(2) Examples and Comparative Examples on a nonaqueous electrolyte

[Production of a nonaqueous electrolyte]

· Example 1

The additive (1 mol/kg) represented by the formula 3Z-3 and 1 mol/kg of LiBF<sub>4</sub> were added to 1.2 g of γ-butyrolactone, and these were dissolved in a globe box at 40°C to obtain a nonaqueous electrolyte.

· Examples 2 to 12

Nonaqueous electrolytes were obtained in the same manner as in Example 1 except that types and amounts of additives, salts and aprotic solvents shown in Table 2 were used.

· Comparative Examples 1 to 3

Nonaqueous electrolytes were obtained in the same manner as in Example 1 except that types and amounts of additives, salts and aprotic solvents shown in Table 2 were used.

[Measurement of a lithium ion transport rate]

A polypropylene porous separator having a diameter of 13 mm was dipped with each of the nonaqueous electrolytes obtained in the foregoing Examples and Comparative Examples. After the dipping was conducted satisfactorily, the resulting separator was held between lithium metal electrodes having the same diameter. A lithium ion transport rate was measured

by a DC polarization method. The results are also shown in  
**Table 2.**

Table 2

Example	Additive and its concentration (mol/kg)	Salt and its concentration (mol/kg)	Aprotic solvent	Lithium ion transport rate
1	3Z-3	1 LiBF <sub>4</sub>	1 GBL	0.77
2	3Z-13	0.03 LiPF <sub>6</sub>	0.01 PC	0.66
3	4Z-3	0.05 LiClO <sub>4</sub>	0.1 PC:DEC=1:1	0.70
4	5Z-1	0.01 LiAsF <sub>6</sub>	10 PC:DME=1:2	0.67
5	6Z-1	0.005 LiCF <sub>3</sub> SO <sub>3</sub>	5 EC:GBL=3:7	0.76
6	3Z-3	0.05 LiN(CF <sub>3</sub> SO <sub>2</sub> ) <sub>2</sub>	0.05 EC:EMC=2:1	0.73
7	3Z-13	2 LiN(C <sub>2</sub> F <sub>5</sub> SO <sub>2</sub> ) <sub>2</sub>	2 EC:DEC:DMC=1:1:1	0.71
8	4Z-3	8 Li(CF <sub>3</sub> SO <sub>2</sub> ) <sub>3</sub>	8 PC:DEC=1:1	0.73
9	5Z-1	0.005 LiCl	0.5 PC:DME=1:2	0.71
10	6Z-1	0.03 LiF	3 EC:DO=1:1	0.74
11	3Z-3	0.3 LiBr	0.3 EC:SL=1:2	0.72
12	3Z-13	6 LiI	6 EC:DME=1:2	0.74
Comparative Example				
1	4Z-3	0.001 LiN(CF <sub>3</sub> SO <sub>2</sub> ) <sub>2</sub>	0.001 EC:GBL=3:7	0.50
2	5Z-1	15 LiCl	15 EC:EMC=2:1	unmeasurable
3	not added	LiBF <sub>4</sub>	1 EC:DEC:DMC=1:1:1	0.30

\* EC: ethylene carbonate, GBL:  $\gamma$ -butyrolactone, DO: 1,3-dioxolane, DME: 1,2-dimethoxyethane, SL:sulfolane  
 PC: propylene carbonate, DEC: diethyl carbonate, EMC: ethyl methyl carbonate, DMC: dimethyl carbonate

## **Industrial Applicability**

The polymeric electrolyte and the nonaqueous electrolyte of the invention can accelerate dissociation of an electrolytic salt upon using a compound having plural trivalent boron atoms in the structure, a Lewis acid, as an additive and consequently improve a transport rate of charge carrier ions. Besides, since an effect of addition per unit weight is great, the transport rate can be improved much, and an effective concentration region of the additive is also wide.

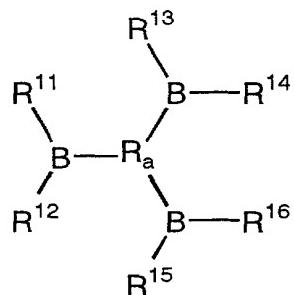
Accordingly, the use of them can provide cells having a higher voltage and a higher capacity than usual ones. Although the usage of cells is not limited, they are preferably used in portable electric appliances such as a video, a camera, a personal computer, a cellular phone and the like.

CLAIMS

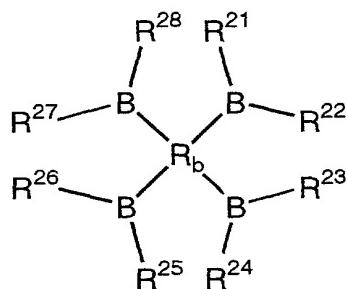
1. A polymeric electrolyte made of an electrolytic salt and a polymeric compound forming a complex with the electrolytic salt, characterized by containing a compound having boron atoms in the structure.

2. The polymeric electrolyte according to claim 1, characterized in that the compound having boron atoms in the structure is one or more selected from the group consisting of compounds represented by the following general formulas (1) to (4).

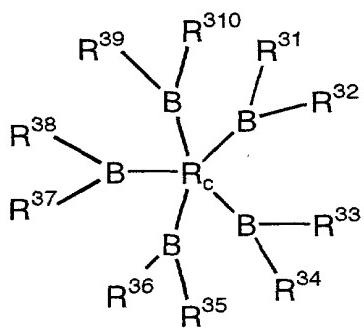
general formula (1)



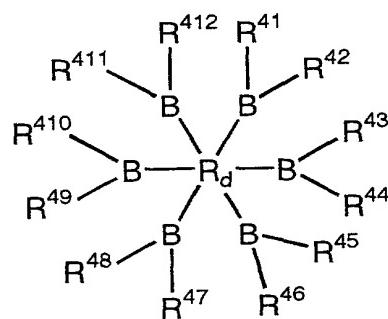
general formula (2)



general formula (3)



general formula (4)



wherein

$R^{11}$ ,  $R^{12}$ ,  $R^{13}$ ,  $R^{14}$ ,  $R^{15}$  and  $R^{16}$  in formula (1),  $R^{21}$ ,  $R^{22}$ ,  $R^{23}$ ,  $R^{24}$ ,  $R^{25}$ ,  $R^{26}$ ,  $R^{27}$  and  $R^{28}$  in formula (2),  $R^{31}$ ,  $R^{32}$ ,  $R^{33}$ ,  $R^{34}$ ,  $R^{35}$ ,  $R^{36}$ ,  $R^{37}$ ,  $R^{38}$ ,  $R^{39}$  and  $R^{310}$  in formula (3), and  $R^{41}$ ,  $R^{42}$ ,  $R^{43}$ ,  $R^{44}$ ,  $R^{45}$ ,  $R^{46}$ ,  $R^{47}$ ,  $R^{48}$ ,  $R^{49}$ ,  $R^{410}$ ,  $R^{411}$  and  $R^{412}$  in formula (4), which may be the same or different, each represent a hydrogen atom, a halogen atom or a monovalent group, or are bound to each other to form a ring,

$R_a$  in formula (1) represents a group having a site capable of being bound to at least 3 boron atoms which are the same or different,  $R_b$  in formula (2) represents a group having a site capable of being bound to at least 4 boron atoms which are the same or different,  $R_c$  in formula (3) represents a group having a site capable of being bound to at least 5 boron atoms which are the same or different, and  $R_d$  in formula (4) represents a group having a site capable of being bound to at least 6 boron atoms which are the same or different.

3. The polymeric electrolyte according to claim 2, characterized in that  $R^{11}$ ,  $R^{12}$ ,  $R^{13}$ ,  $R^{14}$ ,  $R^{15}$ ,  $R^{16}$ ,  $R^{21}$ ,  $R^{22}$ ,  $R^{23}$ ,  $R^{24}$ ,  $R^{25}$ ,  $R^{26}$ ,  $R^{27}$ ,  $R^{28}$ ,  $R^{31}$ ,  $R^{32}$ ,  $R^{33}$ ,  $R^{34}$ ,  $R^{35}$ ,  $R^{36}$ ,  $R^{37}$ ,  $R^{38}$ ,  $R^{39}$ ,  $R^{310}$ ,  $R^{41}$ ,  $R^{42}$ ,  $R^{43}$ ,  $R^{44}$ ,  $R^{45}$ ,  $R^{46}$ ,  $R^{47}$ ,  $R^{48}$ ,  $R^{49}$ ,  $R^{410}$ ,  $R^{411}$  and  $R^{412}$  are one or more groups selected from the member consisting of an alkyl group, an aryl group and fluorine-substituted derivatives thereof.

4. The polymeric electrolyte according to any one of claims 1 to 3, characterized in that the polymeric compound is one or more selected from the group consisting of a polyalkylene, a polyether, a polyester, a polyamine, a polyimide, a polyurethane, a polysulfide, a polyphosphazene, a polysiloxane, derivatives thereof, copolymers thereof and crosslinked products thereof.

5. The polymeric electrolyte according to any one of claims 1 to 3, characterized in that the polymeric compound is one or more selected from the group consisting of a polyalkylene oxide, polyvinylidene fluoride, polyhexafluoropropylene, polyacrylonitrile, polymethyl methacrylate, derivatives thereof, copolymers thereof and crosslinked products thereof.

6. The polymeric electrolyte according to any one of claims 1 to 5, characterized in that the electrolytic salt is a metallic salt.

7. The polymeric electrolyte according to claim 6, characterized in that the metallic salt is a lithium salt.

8. The polymeric electrolyte according to claim 7, characterized in that the lithium salt is one or more selected from the group consisting of  $\text{LiBF}_4$ ,  $\text{LiPF}_6$ ,  $\text{LiClO}_4$ ,  $\text{LiAsF}_6$ ,  $\text{LiCF}_3\text{SO}_3$ ,  $\text{LiN}(\text{CF}_3\text{SO}_2)_2$ ,  $\text{LiN}(\text{C}_2\text{F}_5\text{SO}_2)_2$ ,  $\text{LiC}(\text{CF}_3\text{SO}_2)_3$ ,  $\text{LiCl}$ ,  $\text{LiF}$ ,  $\text{LiBr}$ ,  $\text{LiI}$ , derivatives thereof and like.

9. The polymeric electrolyte according to claims 1

to 8, characterized by further containing a nonaqueous solvent.

10. The polymeric electrolyte according to claim 9, characterized in that the nonaqueous solvent is an aprotic solvent.

11. The polymeric electrolyte according to claim 10, characterized in that the aprotic solvent is one or more selected from the group consisting of carbonates, lactones, ethers, sulfolanes and dioxolanes.

12. The polymeric electrolyte according to claims 1 to 11, characterized in that the molar ratio of the compound having boron atoms in the structure to the electrolytic salt is 0.1:100 to 300:100.

13. An electric device using the polymeric electrolyte according to any one of claims 1 to 12.

14. A cell in which a positive electrode and a negative electrode are linked through the polymeric electrolyte according to any one of claims 1 to 12.

15. The cell according to claim 14, characterized in that the positive electrode is made of a double metal oxide capable of occluding and releasing lithium ions, and the negative electrode is made of a lithium metal, a lithium alloy or a compound capable of reversibly occluding and releasing lithium ions.

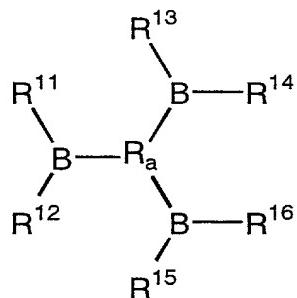
16. A nonaqueous electrolyte made of an electrolytic

salt and a nonaqueous solvent that dissolves the electrolytic salt, characterized by containing a compound having boron atoms in the structure.

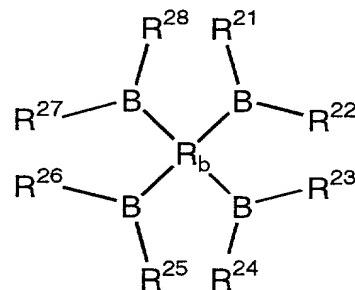
17. The nonaqueous electrolyte according to claim 16, characterized in that the compound having boron atoms in the structure is one or more selected from the group consisting of compounds represented by the following general formulas

(1) to (4)

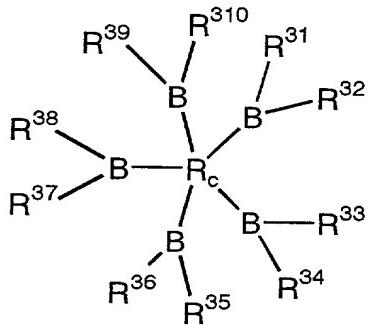
general formula (1)



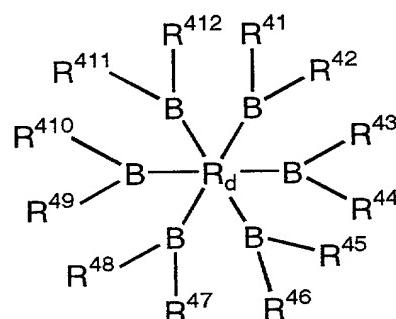
general formula (2)



general formula (3)



general formula (4)



wherein

R<sup>11</sup>, R<sup>12</sup>, R<sup>13</sup>, R<sup>14</sup>, R<sup>15</sup> and R<sup>16</sup> in formula (1), R<sup>21</sup>, R<sup>22</sup>, R<sup>23</sup>, R<sup>24</sup>, R<sup>25</sup>, R<sup>26</sup>, R<sup>27</sup> and R<sup>28</sup> in formula (2), R<sup>31</sup>, R<sup>32</sup>, R<sup>33</sup>, R<sup>34</sup>, R<sup>35</sup>, R<sup>36</sup>, R<sup>37</sup>, R<sup>38</sup>, R<sup>39</sup> in formula (3), and R<sup>41</sup>, R<sup>42</sup>, R<sup>43</sup>, R<sup>44</sup>, R<sup>45</sup>, R<sup>46</sup>, R<sup>47</sup>, R<sup>48</sup>, R<sup>49</sup>, R<sup>410</sup>, R<sup>411</sup> and R<sup>412</sup> in formula (4).

R<sup>35</sup>, R<sup>36</sup>, R<sup>37</sup>, R<sup>38</sup>, R<sup>39</sup> and R<sup>310</sup> in formula (3), and R<sup>41</sup>, R<sup>42</sup>, R<sup>43</sup>, R<sup>44</sup>, R<sup>45</sup>, R<sup>46</sup>, R<sup>47</sup>, R<sup>48</sup>, R<sup>49</sup>, R<sup>410</sup>, R<sup>411</sup> and R<sup>412</sup> in formula (4), which may be the same or different, each represent a hydrogen atom, a halogen atom or a monovalent group, or are bound to each other to form a ring,

Ra in formula (1) represents a group having a site capable of being bound to at least 3 boron atoms which are the same or different, Rb in formula (2) represents a group having a site capable of being bound to at least 4 boron atoms which are the same or different, Rc in formula (3) represents a group having a site capable of being bound to at least 5 boron atoms which are the same or different, and Rd in formula (4) represents a group having a site capable of being bound to at least 6 boron atoms which are the same or different.

18. The nonaqueous electrolyte according to claim 17, characterized in that R<sup>11</sup>, R<sup>12</sup>, R<sup>13</sup>, R<sup>14</sup>, R<sup>15</sup>, R<sup>16</sup>, R<sup>21</sup>, R<sup>22</sup>, R<sup>23</sup>, R<sup>24</sup>, R<sup>25</sup>, R<sup>26</sup>, R<sup>27</sup>, R<sup>28</sup>, R<sup>31</sup>, R<sup>32</sup>, R<sup>33</sup>, R<sup>34</sup>, R<sup>35</sup>, R<sup>36</sup>, R<sup>37</sup>, R<sup>38</sup>, R<sup>39</sup>, R<sup>310</sup>, R<sup>41</sup>, R<sup>42</sup>, R<sup>43</sup>, R<sup>44</sup>, R<sup>45</sup>, R<sup>46</sup>, R<sup>47</sup>, R<sup>48</sup>, R<sup>49</sup>, R<sup>410</sup>, R<sup>411</sup> and R<sup>412</sup> are one or more groups selected from the member consisting of an alkyl group, an aryl group and fluorine-substituted derivatives thereof.

19. The nonaqueous electrolyte according to any one of claims 16 to 18, characterized in that the electrolytic salt is a metallic salt.

20. The nonaqueous electrolyte according to claim 19, characterized in that the metallic salt is a lithium salt.

21. The nonaqueous electrolyte according to claim 20, characterized in that the lithium salt is one or more selected from the group consisting of LiBF<sub>4</sub>, LiPF<sub>6</sub>, LiClO<sub>4</sub>, LiAsF<sub>6</sub>, LiCF<sub>3</sub>SO<sub>3</sub>, LiN(CF<sub>3</sub>SO<sub>2</sub>)<sub>2</sub>, LiN(C<sub>2</sub>F<sub>5</sub>SO<sub>2</sub>)<sub>2</sub>, LiC(CF<sub>3</sub>SO<sub>2</sub>)<sub>3</sub>, LiCl, LiF, LiBr, LiI, derivatives thereof and the like.

22. The nonaqueous electrolyte according to claims 16 to 21, characterized in that the nonaqueous solvent is an aprotic solvent.

23. The nonaqueous electrolyte according to claim 22, characterized in that the aprotic solvent is one or more selected from the group consisting of carbonates, lactones, ethers, sulfolanes and dioxolanes.

24. The nonaqueous electrolyte according to any one of claims 16 to 23, characterized in that the molar ratio of the compound having the boron atoms in the structure to the electrolytic salt is 0.1:100 to 300:100.

25. An electric device using the nonaqueous electrolyte according to any one of claims 16 to 24.

26. A cell in which a positive electrode and a negative electrode are linked through the nonaqueous electrolyte according to any one of claims 16 to 24 and a separator.

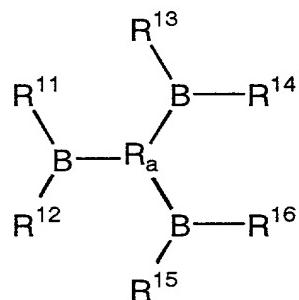
27. The cell according to claim 26, characterized in that the positive electrode is made of a double metal oxide

capable of occluding and releasing lithium ions, and the negative electrode is made of a lithium metal, a lithium alloy or a compound capable of reversibly occluding and releasing lithium ions.

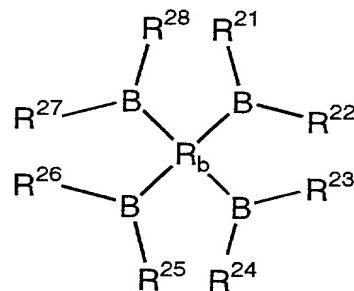
ABSTRACT

Provided are a polymeric electrolyte or a nonaqueous electrolyte that can improve a transport rate of charge carrier ions by adding a compound having boron atoms in the structure, preferably one or more selected from the group consisting of compounds represented by the following general formulas (1) to (4), and an electric device such as a cell or the like using the same.

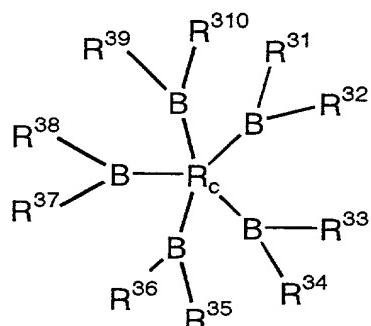
general formula (1)



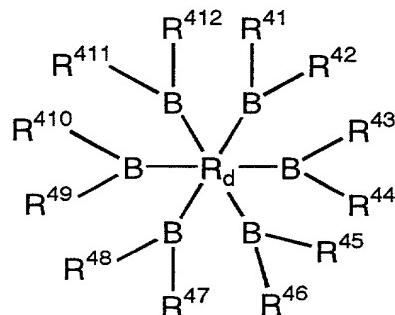
general formula (2)



general formula (3)



general formula (4)



wherein

R<sup>11</sup>, R<sup>12</sup>, R<sup>13</sup>, R<sup>14</sup>, R<sup>15</sup>, R<sup>16</sup>, R<sup>21</sup>, R<sup>22</sup>, R<sup>23</sup>, R<sup>24</sup>, R<sup>25</sup>, R<sup>26</sup>, R<sup>27</sup>,

$R^{28}$ ,  $R^{31}$ ,  $R^{32}$ ,  $R^{33}$ ,  $R^{34}$ ,  $R^{35}$ ,  $R^{36}$ ,  $R^{37}$ ,  $R^{38}$ ,  $R^{39}$ ,  $R^{310}$ ,  $R^{41}$ ,  $R^{42}$ ,  $R^{43}$ ,  $R^{44}$ ,  $R^{45}$ ,  $R^{46}$ ,  $R^{47}$ ,  $R^{48}$ ,  $R^{49}$ ,  $R^{410}$ ,  $R^{411}$  and  $R^{412}$  each represent a hydrogen atom, a halogen atom or a monovalent group, or represent groups bound to each other to form a ring, and  $R_a$ ,  $R_b$ ,  $R_c$  and  $R_d$  each represent a group having a site capable of being bound to boron atoms which are the same or different.

**COMBINED DECLARATION FOR PATENT APPLICATION AND  
POWER OF ATTORNEY**  
(Includes Reference to PCT International Applications)

Attorney's Docket Number

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name,

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

**POLYMERIC ELECTROLYTE, NONAQUEOUS ELECTROLYTE**

**AND ELECTRIC DEVICE USING THE SAME**

the specification of which (check only one item below):

- [ ] is attached hereto.
- [X] was filed as United States application  
Serial No. 09/787,231  
on March 16, 2001,  
and was amended  
on \_\_\_\_\_ (if applicable).
- [X] was filed as PCT international application  
Number PCT/JPOO/05812  
on August 28, 2000  
and was amended under PCT Article 19  
on \_\_\_\_\_ (if applicable).

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to the examination of this application in accordance with Title 37, Code of Federal Regulations, §1.56(a).

I hereby claim foreign priority benefits under Title 35, United States Code, §119 of any foreign application(s) for patent or inventor's certificate or of any PCT international application(s) designating at least one country other than the United States of America listed below and have also identified below any foreign application(s) for patent or inventor's certificate or any PCT international application(s) designating at least one country other than the United States of America filed by me on the same subject matter having a filing date before that of the application(s) of which priority is claimed:

**PRIOR FOREIGN/PCT APPLICATION(S) AND ANY PRIORITY CLAIMS UNDER 35 U.S.C. 119:**

Country (if PCT indicate "PCT")	Application Number	Date of Filing (day, month, year)	Priority Claimed Under 35 USC 119
Japan	11-248890	02/09/1999	[X] Yes [ ] No
Japan	11-248891	02/09/1999	[X] Yes [ ] No
			[ ] Yes [ ] No
			[ ] Yes [ ] No
			[ ] Yes [ ] No

**COMBINED DECLARATION FOR PATENT APPLICATION AND  
POWER OF ATTORNEY (Continued)**  
(Includes Reference to PCT International Applications)

Attorney's Docket Number

I hereby claim the benefit under Title 35, United States Code, §120 of any United States application(s) or PCT international application(s) designating the United States of America that is/are listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in that/those prior application(s) in the manner provided by the first paragraph of Title 35, United States Code, §112, I acknowledge the duty to disclose material information as defined in Title 37, Code of Federal Regulations, §1.56(a) which occurred between the filing date of the prior application(s) and the national or PCT international filing date of this application:

**PRIOR U.S. APPLICATIONS OR PCT INTERNATIONAL APPLICATIONS DESIGNATING THE U.S. FOR BENEFIT UNDER  
35 U.S.C. 120:**

U.S. APPLICATIONS		STATUS (Check One)		
U.S. Application Number	U. S. Filing Date	Patented	Pending	Abandoned
<b>PCT APPLICATIONS DESIGNATING THE U.S.</b>				
PCT Application No.	PCT Filing Date	U.S. Serial Numbers Assigned (if any)		
PCT/JPOO/05812	August 28, 2000	09/787,231		

**POWER OF ATTORNEY:** As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith.

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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

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